Supporting Information to accompany

Correlating Luminescence and Single-Molecule Magnetism for Two Series of Heteroleptic Lanthanoid Complexes

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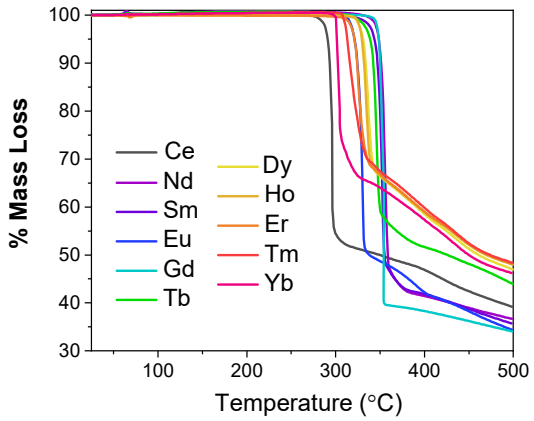

Fig. S1 Thermogravimetric analysis (TGA) between 25 - 500°C under N_2 flow for **1-Ln** demonstrating thermal stability in the solid state.

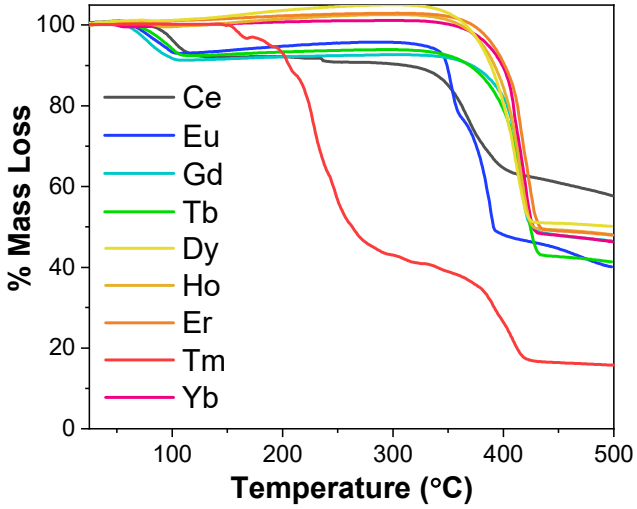

Fig. S2 Thermogravimetric analysis (TGA) between 25 - 500°C under N_2 flow of **2-Ln** demonstrating thermal stability in the solid state.

Table S1 Crystallographic data and structure refinement parameters for compounds **1-Ce**, **1-Nd**, **1-Sm**, **1-Gd** and **1-Tb**.

	1-Ce	**1-Nd**	**1-Sm**	**1-Gd**	**1-Tb**
Empirical formula	$C_{20}H_{21}CeN_8O_9$	$C_{20}H_{21}NdN_8O_9$	$C_{20}H_{21}SmN_8O_9$	$C_{20}H_{21}GdN_8O_9$	$C_{20}H_{21}TbN_8O_9$
Formula weight	657.57	661.69	667.80	674.70	676.37
Temperature / K	100.15	100.00(10)	100.00(10)	100.00(10)	100.00(10)
Crystal system	Monoclinic	Monoclinic	Monoclinic	Monoclinic	Monoclinic
Space group	C2/c	C2/c	C2/c	C2/c	C2/c
a / Å	21.9783(3)	21.9161(4)	21.8885(3)	21.8507(3)	21.8449(3)
b / Å	8.45040(10)	8.41440(10)	8.37110(10)	8.34760(10)	8.31870(10)
c / Å	27.0691(4)	27.0806(5)	27.1008(3)	27.1171(3)	27.1436(4)
α / °	90	90	90	90	90
β / °	99.9270(10)	99.914(2)	99.8490(10)	9.8200(10)	99.773(2)
γ / °	90	90	90	90	90
Volume / Å^3	4952.15(12)	4919.38(14)	4892.50(11)	4873.71(10)	4860.99(12)
Z	8	8	8	8	8
ρcalc / g cm^{-3}	1.764	1.787	1.813	1.839	1.848
μ / mm-1	14.803	16.706	18.630	18.209	14.916
F(000)	2616.0	2632.0	2648.0	2664.0	2672.0
Crystal size / mm^3	0.639×0.048×0.041	0.581×0.049×0.032	0.16×0.034×0.028	0.417×0.036×0.028	0.407×0.078×0.028
Radiation	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)
2θ range / °	8.168 to 160.278	6.626 to 160.784	6.62 to 160.966	6.616 to 160.696	6.608 to 160.318
Index ranges	−28≤h≤27 −10≤k≤9 −33≤l≤34	−27≤h≤28 −10≤k≤8 −34≤l≤34	−26≤h≤27 −10≤k≤10 −34≤l≤34	−27≤h≤27 −10≤k≤9 −33≤l≤34	−27≤h≤27 −10≤k≤9 −33≤l≤34
Reflections collected	38431	29582	30830	31860	32459
Independent reflections	5353 [R_{int} = 0.0679, R_{sigma} = 0.0311]	5313 [R_{int} = 0.0998, R_{sigma} = 0.0952]	5289 [R_{int} = 0.0667, R_{sigma} = 0.0427]	5249 [R_{int} = 0.0683, R_{sigma} = 0.0419]	5232 [R_{int} = 0.0607, R_{sigma} = 0.0360]
Data/restraints/parameters	5353/0/344	5313/0/345	5289/0/345	5249/0/345	5232/0/345
Goodness-of-fit on F2	1.112	1.113	1.086	1.100	1.120
Final R indexes [I ≥ 2σ (I)]	R1 = 0.0357, wR2 = 0.1032	R1 = 0.0511, wR2 = 0.1386	R1 = 0.0281, wR2 = 0.0721	R1 = 0.0349, wR2 = 0.0904	R_1 = 0.0380, wR_2 = 0.1077
Final R indexes [all data]	R1 = 0.0368, wR2 = 0.1042	R1 = 0.0597, wR2 = 0.1415	R1 = 0.0299, wR2 = 0.0730	R1 = 0.0385, wR2 = 0.0926	R_1 = 0.0420, wR_2 = 0.1101
Largest diff. peak/hole / eÅ^{-3}	1.41/−1.18	2.37/−2.14	0.63/−0.59	0.73/−0.68	0.84/−0.86

Table S2 Crystallographic data and structure refinement parameters for compounds **1-Dy**, **1-Er**, **1-Tm** and **1-Yb**.

	1-Dy	**1- Er**	**1-Tm**	**1-Yb**
Empirical formula	$C_{20}H_{21}DyN_8O_9$	$C_{20}H_{21}ErN_8O_9$	$C_{20}H_{21}TmN_8O_9$	$C_{20}H_{21}N_8O_9Yb$
Formula weight	679.95	684.71	686.38	690.49
Temperature / K	100.00(10)	100.00(10)	100.00(10)	129.99(10)
Crystal system	Monoclinic	Monoclinic	Monoclinic	monoclinic
Space group	C2/c	C2/c	C2/c	C2/c
a / Å	21.8229(2)	21.7876(2)	21.7771(2)	21.7909(3)
b / Å	8.31060(10)	8.28920(10)	8.26530(10)	8.27130(10)
c / Å	27.1281(3)	27.1254(2)	27.1649(2)	27.1978(3)
α / º	90	90	90	90
β / º	99.7770(10)	99.7860(10)	99.6960(10)	99.6980(10)
γ / º	90	90	90	90
Volume / Å^3	4848.53(9)	4827.61(8)	4819.68(8)	4832.05(10)
Z	8	8	8	8
ρcalc / g cm^{-3}	1.863	1.884	1.892	1.898
μ / mm^{-1}	17.099	7.065	7.508	7.77
F(000)	2680.0	2696.0	2704.0	2712
Crystal size / mm^3	0.306x0.04x0.036	0.643 ×0.062×0.055	0.982×0.057×0.048	0.51 × 0.08 × 0.07
Radiation	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)
2θ range / º	6.612 to 160.164	6.614 to 160.43	6.602 to 160.41	6.594 to 160.026
Index ranges	−27≤h≤27 −9≤k≤10 −34≤l≤34	-27≤h≤27 -8≤k≤10 -33≤l≤34	-27≤h≤27 -9≤k≤10 -33≤l≤34	-27 ≤ h ≤ 26, -10 ≤ k ≤ 7, -34 ≤ l ≤ 31
Reflections collected	31411	30768	31169	17308
Independent reflections	5237 [R_{int} = 0.0490, R_{sigma} = 0.0322]	5228 [R_{int} = 0.0615, R_{sigma} = 0.0474]	5211 [R_{int} = 0.0355, R_{sigma} = 0.0194]	5091 [R_{int} = 0.0462, R_{sigma} = 0.0430]
Data/restraints/parameters	5237/0/345	5228/0/345	5211/0/345	5091/0/345
Goodness-of-fit on F^2	1.105	1.122	1.078	1.094
Final R indexes [I ≥ 2σ (I)]	R_1 = 0.0250, wR_2 = 0.0618	R_1 = 0.0322, wR_2 = 0.0855	R_1 = 0.0272, wR_2 = 0.0780	R_1 = 0.0308, wR_2 = 0.0831
Final R indexes [all data]	R_1 = 0.0275, wR_2 = 0.0628	R_1 = 0.366, wR_2 = 0.0871	R_1 = 0.0278, wR_2 = 0.0785	R_1 = 0.0328, wR_2 = 0.0847
Largest diff. peak/hole / eÅ^{-3}	0.41/-0.41	0.76/-0.95	0.95/-1.19	1.26/-1.02

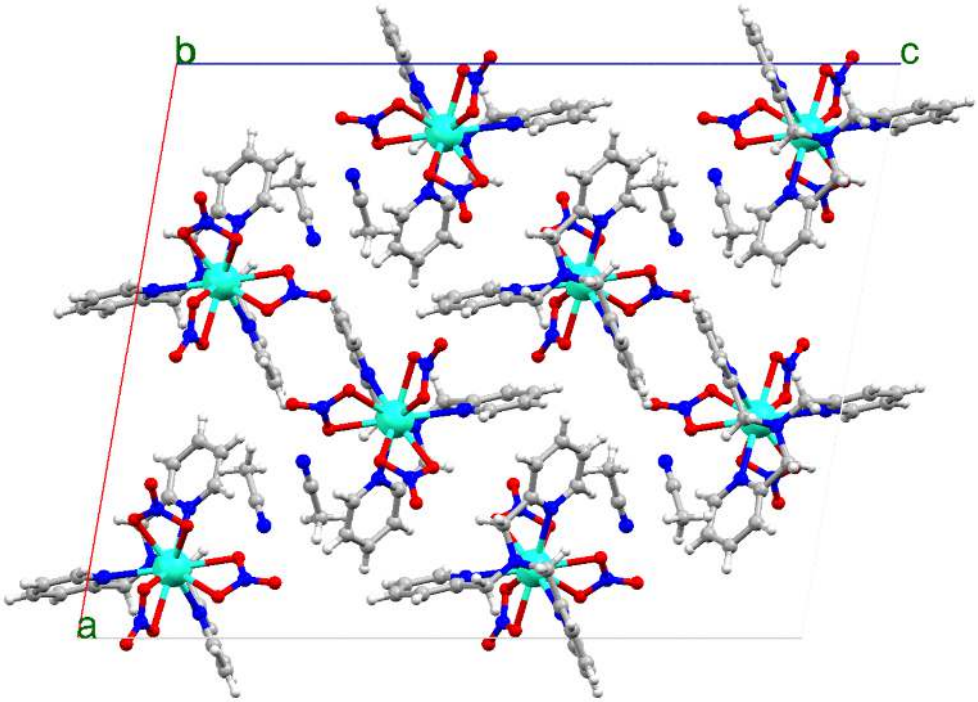

Fig. S3 Depiction of the unit cell of **1-Ln** as viewed along the b-axis, with cell edges shown in blue. Color code: carbon (grey), hydrogen (white), oxygen (red), nitrogen (dark blue), and lanthanoid (cyan).

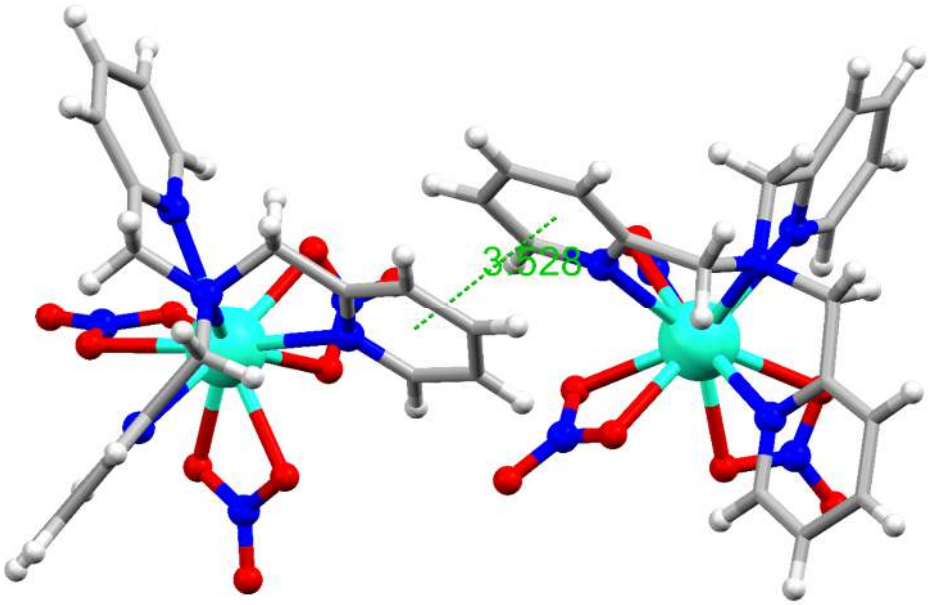

Fig. S4 Depiction of the p××p stacking interaction between two adjacent molecules of **1-Ln** with centroid-to-centroid distance depicted. Color code: carbon (grey), hydrogen (white), oxygen (red), nitrogen (dark blue), and lanthanoid (cyan).

Table S3 Selected interatomic distances (Å) and SHAPE index values for **1-Ln**.

	1-Ce	**1-Nd**	**1-Sm**	**1-Gd**	**1-Tb**	**1-Dy**	**1- Er**	**1-Tm**
Ln1-N1	2.682(3)	2.648(4)	2.613(2)	2.596(3)	2.582(3)	2.565(2)	2.542(2)	2.531(2)
Ln1-N2	2.667(3)	2.625(4)	2.608(2)	2.650(4)	2.638(3)	2.577(2)	2.541(2)	2.610(2)
Ln1-N3	2.665(3)	2.701(4)	2.668(3)	2.589(3)	2.573(3)	2.630(3)	2.567(3)	2.558(2)
Ln1-N4	2.711(3)	2.638(4)	2.615(3)	2.597(4)	2.588(3)	2.557(2)	2.616(3)	2.531(2)
Ln1-O1	2.602(3)	2.576(4)	2.554(2)	2.481(3)	2.472(3)	2.460(2)	2.479(3)	2.479(2)
Ln1-O2	2.559(2)	2.522(3)	2.490(2)	2.464(3)	2.458(3)	2.442(2)	2.417(3)	2.407(2)
Ln1-O3	2.560(3)	2.572(3)	2.544(2)	2.460(3)	2.512(3)	2.504(2)	2.494(3)	2.429(2)
Ln1-O4	2.570(3)	2.518(3)	2.487(2)	2.528(3)	2.449(3)	2.441(2)	2.417(2)	2.417(2)
Ln1-O5	2.602(3)	2.535(4)	2.505(2)	2.464(3)	2.452(3)	2.513(2)	2.422(2)	2.490(2)
Ln1-O6	2.555(3)	2.522(3)	2.498(2)	2.531(3)	2.528(3)	2.438(2)	2.438(2)	2.408(2)
Ln···Ln[a]	7.778(6)	7.7380(7)	7.7246(6)	7.7064(6)	7.7016(7)	7.6924(6)	7.6755(6)	7.6783(6)

[a]Closest intermolecular distance. [b]*SHAPE* index for sphenocorona geometry (10-coordinate) in *SHAPE 2.1*.

Table S4 SHAPE parameters for **1-Ln** for 10-coordinate in SHAPE 2.1.

	1-Ce	**1-Nd**	**1-Sm**	**1-Gd**	**1-Tb**	**1-Dy**	**1- Er**	**1-Tm**
EPY-10	25.209	25.216	25.185	25.153	25.148	25.154	25.176	25.094
OBPY-10	17.491	17.417	17.434	17.401	17.295	17.301	17.238	17.237
PPR-10	8.699	8.844	8.924	8.941	9.015	9.023	9.08	9.097
PAPR-10	13.907	13.866	13.833	13.866	13.854	13.873	13.911	13.895
JBCCU-10	14.716	14.581	14.461	14.423	14.378	14.335	14.313	14.246
JBCSAPR-10	7.024	6.863	6.709	6.572	6.481	6.448	6.323	6.251
JMBIC-10	9.739	9.724	9.745	9.813	9.82	9.849	9.894	9.881
JATDI-10	16.167	16.337	16.524	16.669	16.783	16.818	16.929	17.002
JSPC-10	3.405	3.179	3.02	2.897	2.829	2.776	2.671	2.606
SDD-10	7.796	7.61	7.418	7.351	7.264	7.246	7.181	7.106
TD-10	7.711	7.481	7.284	7.196	7.103	7.074	6.986	6.906

EPY-10 = enneagonal pyramid, OBPY-10 = octagonal bipyramid, PPR-10 = pentagonal prism, PAPR-10 = pentagonal antiprism, JBCCU-10 = bicapped cube, JBCSAPR-10 = bicapped square antiprism, JMBIC-10 = metabidiminished icoahedron, JATDI-10 = augmented tridiminished icosahedron, JSPC-10 = sphenocorona, SDD-10 = staggered dodecahedron, TD-10 = tetradecahedron, HD-10 = hexadecahedron.

Table S5 CSoM parameters for **1-Ln** including the coordinated atoms only.

	1-Ce	**1-Nd**	**1-Sm**	**1-Gd**	**1-Tb**	**1-Dy**	**1- Er**	**1-Tm**
C_2	42.3	42.9	43.7	44.2	44.6	44.8	45.2	45.5
C_{2h}	86.9	86.5	86.1	85.8	85.6	85.5	85.2	85.0
C_{2v}	25.4	25.9	26.4	26.7	27.0	27.1	27.4	27.6
C_3	**0.22**	**0.22**	**0.22**	**0.22**	**0.21**	**0.22**	**0.22**	**0.23**
C_{3h}	72.8	72.7	72.5	72.3	72.3	72.3	72.1	72.0
C_{3v}	2.1	1.9	1.8	1.8	1.7	1.7	1.6	1.6
C_4	25.1	25.3	25.6	25.8	25.9	26.0	26.2	26.3
C_{4h}	86.6	86.5	86.4	85.6	85.3	85.1	84.7	84.3
C_{4v}	21.6	21.7	21.9	22.0	22.1	22.2	22.3	22.4
C_5	23.8	24.1	24.5	24.7	24.9	25.0	25.2	25.3
C_{5h}	82.1	81.9	81.8	81.7	81.7	81.7	81.6	81.5
C_{5v}	21.4	21.7	21.9	22.0	22.1	22.2	22.3	22.4
C_6	25.4	25.8	26.3	26.6	26.8	26.9	27.2	27.4
C_{6h}	72.0	71.7	71.0	70.8	70.6	70.8	70.6	70.2
C_{6v}	20.8	21.2	21.6	21.9	22.1	22.2	22.5	22.6
C_7	21.6	21.8	22.1	22.3	22.4	22.4	22.6	22.7
C_8	22.2	22.3	22.5	22.5	22.6	22.6	22.7	22.8
C_i	134.7	134.0	133.3	132.8	132.5	132.4	132.0	131.5
C_s	3.9	3.5	3.4	3.2	3.1	3.1	2.9	2.9
D_2	93.5	92.9	92.2	91.6	91.4	91.3	90.8	90.5
D_{2d}	83.8	83.2	82.5	82.0	81.7	81.5	81.1	80.8
D_{2h}	82.7	82.7	82.6	82.3	82.1	81.9	81.5	81.1
D_3	70.4	70.2	69.9	69.7	69.6	69.5	69.4	69.1
D_{3d}	71.2	70.9	70.5	70.2	70.1	70.0	69.8	69.5
D_{3h}	79.3	79.0	78.9	78.1	78.0	77.9	77.4	78.2
D_4	84.0	83.5	83.3	82.3	82.3	82.2	82.7	82.8
D_{4d}	76.4	76.2	76.0	75.8	75.8	75.8	75.6	75.5
D_{4h}	82.8	82.3	78.8	78.3	82.3	82.3	82.1	77.1
D_5	80.9	80.8	80.6	80.4	80.4	80.4	80.3	80.1
D_{5d}	77.5	77.3	77.1	77.0	77.0	77.0	76.8	76.7
D_{5h}	77.4	77.2	77.0	76.8	76.8	76.8	76.6	76.5
D_6	71.7	70.8	70.5	71.0	70.9	70.8	70.6	70.4
D_{6h}	78.4	78.1	77.6	77.4	76.9	76.8	76.5	76.7
D_{7h}	52.1	52.1	52.1	52.1	52.1	52.1	52.1	52.1
D_{8h}	76.6	76.4	76.1	76.0	76.1	76.0	75.7	75.6
I	90.7	90.3	89.6	89.7	89.2	89.2	88.8	88.5
I_h	90.8	90.1	89.6	89.3	89.2	88.7	88.6	88.3
O	79.3	78.7	77.9	77.5	77.2	77.1	76.6	76.3
O_h	78.1	77.4	76.7	76.3	76.0	75.9	75.4	75.1
S_{10}	98.1	97.8	97.5	97.3	97.0	96.8	96.4	96.1
S_4	96.4	95.2	94.6	94.0	93.0	92.8	92.2	91.7
S_6	80.5	80.1	79.6	79.2	79.1	79.0	78.7	78.4
S_8	83.3	83.1	82.9	82.8	82.7	82.7	82.6	82.4

T	81.3	81.3	79.5	79.2	78.6	78.3	77.8	77.4
T_d	77.7	76.5	75.7	75.1	74.8	74.7	74.2	73.8
T_h	80.4	79.7	79.0	78.5	78.2	78.0	77.6	77.3

Table S6 CSoM parameters for **1-Ln** including all atoms.

	1-Ce	**1-Nd**	**1-Sm**	**1-Gd**	**1-Tb**	**1-Dy**	**1- Er**	**1-Tm**
C_2	54.5	53.9	54.2	54.2	53.8	54.0	53.7	53.7
C_{2h}	139.2	139.3	134.9	133.5	131.9	131.2	135.2	129.2
C_{2v}	33.9	33.9	33.9	34.2	34.0	34.1	34.1	34.1
C_3	**0.60**	**0.60**	**0.61**	**0.61**	**0.60**	**0.62**	**0.62**	**0.62**
C_{3h}	120.4	117.7	116.1	114.9	113.8	113.1	112.0	111.4
C_{3v}	4.0	4.1	4.1	4.1	4.1	4.2	4.2	4.2
C_4	31.7	31.6	31.6	31.6	31.6	31.6	31.6	31.6
C_{4h}	124.3	122.6	121.6	119.7	118.4	118.1	117.2	115.6
C_{4v}	26.8	26.8	26.8	26.8	26.8	26.8	26.8	26.8
C_5	28.4	28.5	28.5	28.5	28.6	28.6	28.6	28.6
C_{5h}	124.8	123.4	120.9	119.9	118.8	118.2	117.3	117.3
C_{5v}	25.4	25.7	25.6	25.6	25.7	25.7	25.7	25.7
C_6	35.5	35.5	35.5	35.6	35.6	35.5	35.5	35.5
C_{6h}	103.3	101.6	99.8	99.0	97.8	96.9	96.1	96.3
C_{6v}	28.1	28.2	28.2	28.3	28.3	28.3	28.4	28.4
C_7	26.7	26.7	26.8	26.8	26.9	26.9	26.9	26.9
C_8	26.4	26.4	26.4	26.4	26.4	26.4	26.4	26.4
C_i	226.9	222.1	219.4	217.2	215.0	213.9	211.9	210.7
C_s	7.2	7.3	7.5	7.6	7.5	7.6	7.7	7.8
D_2	134.0	129.8	128.6	131.9	127.5	130.5	129.0	126.3
D_{2d}	119.0	116.0	117.1	116.3	115.0	114.7	114.8	109.7
D_{2h}	122.5	119.9	123.3	121.5	117.7	116.4	115.6	118.6
D_3	112.3	109.9	107.3	106.3	105.1	104.2	102.9	109.7
D_{3d}	114.2	110.9	110.9	109.7	108.5	108.1	105.1	106.0
D_{3h}	111.5	109.6	108.1	107.0	106.5	106.0	109.5	104.7
D_4	126.4	124.1	120.4	120.3	119.2	121.1	115.9	116.3
D_{4d}	109.3	107.7	105.8	104.3	104.8	108.2	106.8	101.9
D_{4h}	121.5	119.6	115.6	113.1	116.3	116.2	114.3	109.7
D_5	124.4	122.0	120.8	118.1	117.8	118.1	116.4	116.2
D_{5d}	120.2	117.9	116.8	115.8	116.0	114.0	112.6	112.8
D_{5h}	118.7	116.5	114.5	114.1	112.3	111.7	110.5	110.8
D_6	104.0	101.7	99.8	98.7	98.2	97.7	97.3	97.0
D_{6h}	112.4	110.0	109.0	106.8	107.0	105.4	104.5	104.8
D_{7h}	78.4	77.3	76.2	75.6	75.2	75.6	75.0	73.9
D_{8h}	118.4	116.0	113.4	112.4	112.4	111.6	111.0	108.7
I	123.4	120.2	121.1	119.5	118.5	118.1	117.1	116.4
I_h	123.0	120.6	120.6	119.4	116.9	116.6	115.2	114.7
O	114.7	112.7	111.4	110.8	109.6	108.1	107.6	106.7
O_h	111.3	109.5	107.7	107.7	105.8	106.3	105.6	104.5

S_{10}	141.3	138.2	136.5	135.3	134.7	133.4	132.8	131.2
S_4	138.8	133.6	131.7	132.0	128.6	128.3	127.1	127.0
S_6	127.5	124.7	123.2	121.8	120.6	120.0	118.8	118.0
S_8	128.1	125.7	123.6	122.7	121.6	121.4	120.4	119.8
T	118.9	116.3	115.9	114.8	114.6	113.0	112.1	111.9
T_d	115.6	112.6	111.3	110.2	108.7	108.5	106.8	106.0
T_h	115.5	111.6	110.7	108.9	108.6	107.4	107.0	106.9

Table S7 Crystallographic data and structure refinement parameters for compounds **2-Tb**, **2-Dy**, and **2-Ho**.

	2-Tb	**2-Dy**	**2-Ho**
Empirical formula	$C_{18}H_{18}Cl_3N_4Tb$	$C_{18}H_{18}Cl_3N_4Dy$	$C_{18}H_{18}Cl_3N_4Ho$
Formula weight	555.63	559.23	561.64
Temperature / K	100.0(4)	100.00(10)	100.00(10)
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1/c$	$P2_1/c$	$P2_1/c$
a / Å	8.62510(10)	8.60610(10)	8.59630(10)
b / Å	16.23530(10)	16.21160(10)	16.17730(10)
c / Å	14.64480(10)	4.63640(10)	14.63440(10)
α / º	90	90	90
β / º	104.5110(10)	104.4710(10)	104.4010(10)
γ / º	90	90	90
Volume / $Å^3$	1985.31(3)	1977.26(3)	1971.19(3)
Z	4	4	4
ρcalc / g cm^{-3}	1.859	1.879	1.893
μ / mm^{-1}	21.305	24.021	11.311
F(000)	1080.0	1084	1088.0
Crystal size / mm^3	0.51×0.095×0.075	0.278×0.082×0.063	0.217×0.058×0.035
Radiation	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)
2θ range / º	8.28 to 160.666	8.286 to 153.952	8.294 to 160.038
Index ranges	-10≤h≤9 -20≤k≤20 -18≤l≤18	-7≤h≤10 -19≤k≤19 -18≤l≤17	-10≤h≤10 -20≤k≤19 -18≤l≤18
Reflections collected	73355	36893	27371
Independent reflections	4304 [R_{int} = 0.0861, R_{sigma} = 0.0243]	4023 [R_{int} = 0.0763, R_{sigma} = 0.0308]	4278 [R_{int} = 0.0490, R_{sigma} = 0.0272]
Data/restraints/parameters	4304/0/236	4023/0/236	4278/0/236
Goodness-of-fit on F^2	0.838	1.157	1.100
Final R indexes [I ≥ 2σ (I)]	R_1 = 0.0266, wR_2 = 0.0749	R_1 = 0.0329, wR_2 = 0.0903	R_1 = 0.0275, wR_2 = 0.0731
Final R indexes [all data]	R_1 = 0.0368, wR2 = 0.1042	R_1 = 0.0597, wR2 = 0.1415	R_1 = 0.0299, wR2 = 0.0730
Largest diff. peak/hole / $eÅ^{-3}$	1.41/−1.18	2.37/−2.14	0.63/−0.59

Table S8 Crystallographic data and structure refinement parameters for compounds **2- Er**, **2-Tm**, and **2-Yb**.

	2- Er	**2-Tm**	**2-Yb**
Empirical formula	$C_{18}H_{18}Cl_3N_4$ Er	$C_{18}H_{18}Cl_3N_4Tm$	$C_{18}H_{18}Cl_3N_4Yb$
Formula weight	563.97	565.64	569.75
Temperature / K	99.9(3)	99.99(10)	100.01(10)
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1/c$	$P2_1/c$	$P2_1/c$
a / Å	8.58100(10)	8.56790(10)	8.55520(10)
b / Å	16.15580(10)	16.12840(10)	16.11430(10)
c / Å	14.63670(10)	14.62620(10)	14.61710(10)
α / o	90	90	90
β / o	104.3740(10)	104.3090(10)	104.2910(10)
γ / o	90	90	90
Volume / $Å^3$	1965.61(3)	1958.44(3)	1952.77(3)
Z	4	4	4
ρcalc / g cm^{-3}	1.906	1.918	1.938
μ / mm^{-1}	11.750	12.324	12.709
F(000)	1092.0	1096.0	1100.0
Crystal size / mm3	0.496×0.085×0.052	0.294×0.135×0.04	0.261×0.132×0085
Radiation	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)	Cu Kα (λ = 1.54184)
2θ range / º	8.298 to 160.388	8.306 to 160.19	8.31 to 160.388
Index ranges	-9≤h≤10 -20≤k≤20 -18≤l≤18	-10≤h≤10 -19≤k≤20 -18≤l≤18	-10≤h≤10 -20≤k≤20 -18≤l≤18
Reflections collected	26170	26501	25695
Independent reflections	4251 [R_{int} = 0.0385, R_{sigma} = 0.0235]	4245 [R_{int} = 0.0533, R_{sigma} = 0.0298]	4222 [R_{int} = 0.0434, R_{sigma} = 0.0254]
Data/restraints/parameters	4251/0/236	4245/0/235	4222/0/236
Goodness-of-fit on F^2	1.091	1.073	1.149
Final R indexes [I ≥ 2σ (I)]	R_1 = 0.0225, wR_2 = 0.0566	R_1 = 0.0294, wR_2 = 0.0774	R_1 = 0.0278, wR_2 = 0.0724
Final R indexes [all data]	R_1 = 0.0232, wR_2 = 0.0571	R_1 = 0.0300, wR_2 = 0.0779	R_1 = 0.0280, wR_2 = 0.0726
Largest diff. peak/hole / e$Å^{-3}$	0.51/-0.60	0.94/-1.67	1.53/-1.21

Table S9 Selected interatomic distances (Å) and SHAPE index values for **2-Ln**.

	2-Tb	**2-Dy**	**2-Ho**	**2- Er**	**2-Tm**	**2-Yb**
Ln1-N1	2.575(2)	2.560(3)	2.548(2)	2.537(2)	2.528(3)	2.516(3)
Ln1-N2	2.542(2)	2.528(3)	2.487(2)	2.507(2)	2.463(3)	2.453(2)
Ln1-N3	2.520(2)	2.530(3)	2.523(2)	2.475(2)	2.503(3)	2.487(3)
Ln1-N4	2.552(2)	2.505(3)	2.519(2)	2.513(2)	2.498(2)	2.487(2)
Ln1-Cl1	2.6370(7)	2.6174(8)	2.6075(5)	2.5966(7)	2.5901(7)	2.5802(7)
Ln1-Cl2	2.6284(7)	2.6276(9)	2.6147(6)	2.5984(6)	2.5941(7)	2.5863(7)
Ln1-Cl3	2.6229(9)	2.613(1)	2.6061(7)	2.6049(6)	2.586(1)	2.5771(9)
Ln···Ln[a]	8.0426(4)	8.0285(4)	8.0194(4)	8.0116(4)	7.9997(4)	7.9903(4)

[a]Closest intermolecular distance.

Table S10 SHAPE parameters for **2-Ln** for 7-coordinate in SHAPE 2.1.

	2-Tb	**2-Dy**	**2-Ho**	**2- Er**	**2-Tm**	**2-Yb**
HP-7	35.557	35.553	35.585	35.534	35.51	35.531
HPY-7	21.019	21.023	20.887	20.879	20.846	20.821
PBPY-7	5.804	5.76	5.732	5.67	5.659	5.643
COC-7	2.328	2.301	2.254	2.223	2.189	2.153
CTPR-7	2.783	2.779	2.748	2.746	2.724	2.703
JPBPY-7	9.909	9.867	9.792	9.718	9.69	9.687
JETPY-7	19.186	19.127	19.176	19.159	19.115	19.108

HP-7 = heptagon, HPY-7 = Hexagonal pyramid, PBPY-7 = pentagonal bipyramid, COC-7 = capped octahedron, CTPR-7 = capped trigonal prism, JPBPY-7 = Johnson pentagonal bipyramid, JETPY-7 = Johnson elongated triangular pyramid.

Table S11 CSoM parameters for **2-Ln** including the coordinated atoms only.

	2-Tb	**2-Dy**	**2-Ho**	**2- Er**	**2-Tm**	**2-Yb**
C_2	57.8	57.8	57.9	57.9	57.9	58.0
C_{2h}	112.7	112.5	112.0	111.7	111.5	111.3
C_{2v}	30.0	30.2	29.9	30.0	30.1	30.0
C_3	1.1	1.1	1.1	1.2	1.2	1.2
C_{3h}	77.4	77.1	76.7	76.5	76.3	76.1
C_{3v}	**0.86**	**0.88**	**0.88**	**0.91**	**0.93**	**0.92**
C_4	30.2	30.2	30.2	30.3	30.3	30.3
C_{4h}	97.8	97.6	97.2	97.0	96.9	96.7
C_{4v}	24.1	24.1	24.1	24.2	24.2	24.2
C_5	24.8	24.8	24.9	24.9	24.9	24.9
C_{5h}	92.4	92.1	91.7	91.4	91.3	91.1
C_{5v}	22.0	22.1	22.1	22.1	22.1	22.1
C_6	34.8	34.9	34.9	34.9	35.0	35.0
C_{6h}	81.9	81.7	81.3	81.1	80.9	80.7
C_{6v}	24.6	24.6	24.7	24.7	24.7	24.7
C_7	23.3	23.3	23.4	23.4	23.4	23.4
C_8	23.9	23.9	24.0	24.0	24.0	24.0
C_i	179.2	178.7	178.0	177.4	177.2	176.8
C_s	**0.24**	**0.23**	**0.23**	**0.22**	**0.23**	**0.22**
D_2	99.5	99.3	98.7	98.4	98.1	97.9

D_{2d}	85.3	85.1	84.6	84.3	84.1	83.9
D_{2h}	97.9	97.7	97.0	96.8	96.9	96.5
D_3	77.4	77.2	76.8	76.6	76.4	76.2
D_{3d}	92.3	92.1	91.7	91.5	91.3	91.2
D_{3h}	81.9	81.7	81.2	81.0	80.8	80.6
D_4	96.6	96.4	95.9	95.7	95.5	95.3
D_{4d}	86.1	85.9	85.5	85.4	85.2	85.0
D_{4h}	96.5	93.3	95.8	95.4	95.2	95.0
D_5	92.7	92.6	91.9	92.0	91.8	91.5
D_{5d}	89.7	89.5	89.1	89.0	88.8	88.7
D_{5h}	88.0	87.4	87.4	87.3	87.1	86.9
D_6	84.3	83.5	83.8	83.6	83.0	83.0
D_{6h}	90.5	90.8	89.9	89.7	89.4	89.3
D_{7h}	58.6	58.5	58.3	58.2	58.2	58.0
D_{8h}	86.8	86.6	86.3	86.1	85.9	85.8
I	100.7	100.3	100.4	99.9	99.7	99.5
I_h	100.0	99.5	99.2	99.0	98.8	98.6
O	102.7	102.5	101.1	100.9	100.7	100.5
O_h	100.8	100.5	100.2	99.8	99.5	99.5
S_{10}	114.2	114.3	113.4	113.1	112.8	112.6
S_4	99.0	98.7	98.2	97.9	97.6	97.3
S_6	106.3	106.0	105.6	105.3	105.1	104.9
S_8	96.8	96.6	96.2	96.1	95.9	95.7
T	81.5	81.2	80.8	80.5	80.3	80.1
T_d	77.9	77.7	77.2	77.0	76.8	76.6
T_h	101.2	102.2	100.7	101.7	100.1	99.9

Table S12 CSoM parameters for **2-Ln** including all atoms.

	2-Tb	**2-Dy**	**2-Ho**	**2- Er**	**2-Tm**	**2-Yb**
C_2	62.8	62.6	62.4	62.2	62.0	61.6
C_{2h}	192.0	191.0	188.7	188.3	183.4	181.4
C_{2v}	37.4	37.4	37.4	37.5	37.4	37.4
C_3	**0.64**	**0.66**	**0.68**	**0.70**	**0.71**	**0.72**
C_{3h}	179.9	177.4	172.5	170.6	171.1	169.0
C_{3v}	3.9	4.0	4.0	4.0	4.0	4.0
C_4	35.9	35.9	35.9	35.9	35.9	35.9
C_{4h}	169.1	168.2	169.7	165.5	162.8	161.0
C_{4v}	30.1	30.1	30.1	30.1	30.1	30.1
C_5	31.7	31.8	31.7	31.7	31.7	31.7
C_{5h}	170.4	170.0	167.2	163.6	163.1	159.4
C_{5v}	28.3	28.3	28.3	28.3	28.2	28.2
C_6	40.8	40.7	40.7	40.6	40.6	40.6
C_{6h}	137.3	133.7	132.2	132.5	129.0	129.7
C_{6v}	30.9	30.9	30.9	30.9	30.9	30.9
C_7	29.6	29.6	29.6	29.6	29.5	29.5

C_8	29.6	29.7	29.8	29.8	29.9	30.0
C_i	298.5	293.8	289.7	286.1	282.6	285.0
$\mathbf{C_s}$	7.0	7.0	7.0	7.0	7.0	7.0
D_2	180.2	176.2	173.7	172.3	169.6	168.5
D_{2d}	155.7	153.7	149.5	147.9	147.2	144.5
D_{2h}	173.2	168.3	171.6	167.4	167.3	166.2
D_3	180.0	175.6	175.5	173.9	172.2	170.4
D_{3d}	169.0	162.2	161.2	162.1	159.3	156.9
D_{3h}	150.7	155.4	145.0	142.3	139.7	139.3
D_4	178.6	177.7	175.2	173.2	171.3	169.0
D_{4d}	149.2	147.5	145.2	144.0	142.3	141.4
D_{4h}	158.0	159.8	153.0	151.5	151.0	148.1
D_5	175.3	171.2	170.1	167.4	167.2	167.8
D_{5d}	178.6	177.0	174.1	171.6	169.0	167.0
D_{5h}	171.6	166.3	164.7	160.9	159.3	161.2
D_6	144.3	142.2	140.5	139.0	138.6	136.1
D_{6h}	155.6	154.8	150.9	150.3	148.6	146.3
D_{7h}	117.0	116.3	114.6	113.5	112.1	111.2
D_{8h}	167.6	164.8	162.0	160.6	158.8	157.3
I	168.5	167.6	165.4	162.7	161.2	158.6
I_h	168.9	166.8	164.5	162.8	161.0	159.2
O	166.0	166.8	161.5	160.7	158.8	157.0
O_h	163.2	160.5	158.7	156.8	154.9	153.2
S_{10}	191.7	187.7	186.6	183.1	181.7	178.8
S_4	174.0	171.7	169.3	167.6	165.7	164.2
S_6	179.2	175.2	170.3	168.0	165.2	163.3
S_8	179.0	173.8	172.6	170.3	167.6	166.4
T	152.1	148.0	145.6	144.6	142.9	141.7
T_d	143.8	142.7	140.5	138.7	136.4	134.8
T_h	161.7	157.8	159.2	157.8	153.4	152.5

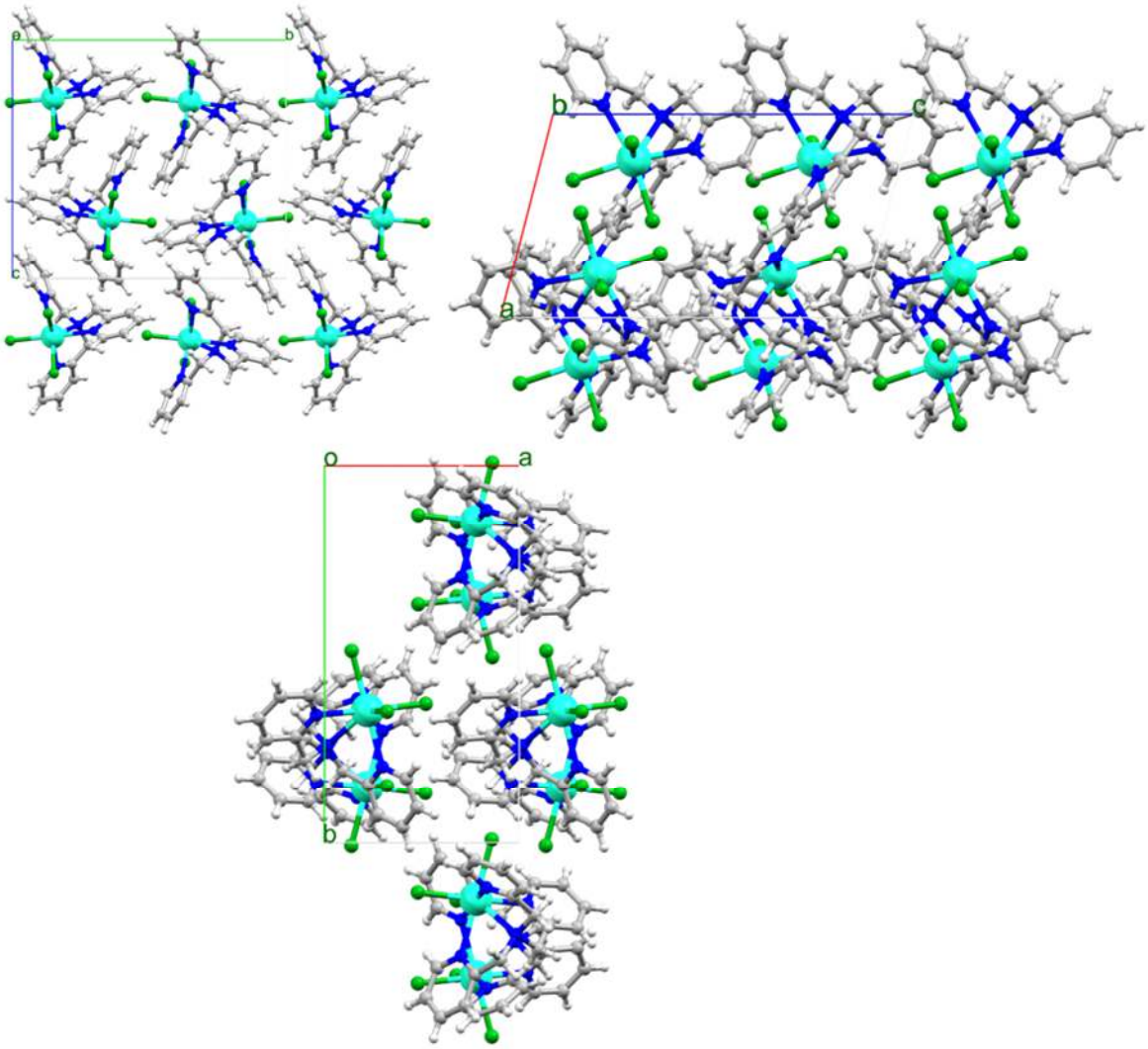

Fig. S5 Depiction of the unit cell of **2-Ln** as viewed along the b-axis, with cell edges shown in green, red, and blue. Color code represents carbon (grey), hydrogen (white), nitrogen (dark blue), chlorine (green) and lanthanoid (cyan).

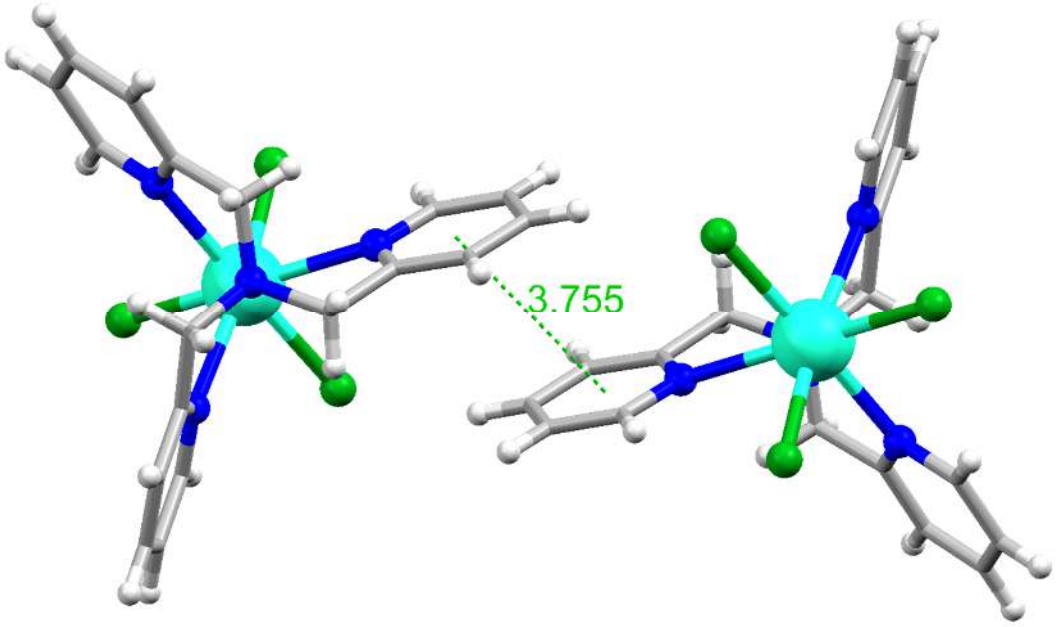

Fig. S6 Depiction of the p-p stacking interaction between two adjacent molecules of **2-Ln** with centroid to centroid distance depicted. Color code represents carbon (grey), hydrogen (white), nitrogen (dark blue), chlorine (green) and lanthanoid (cyan).

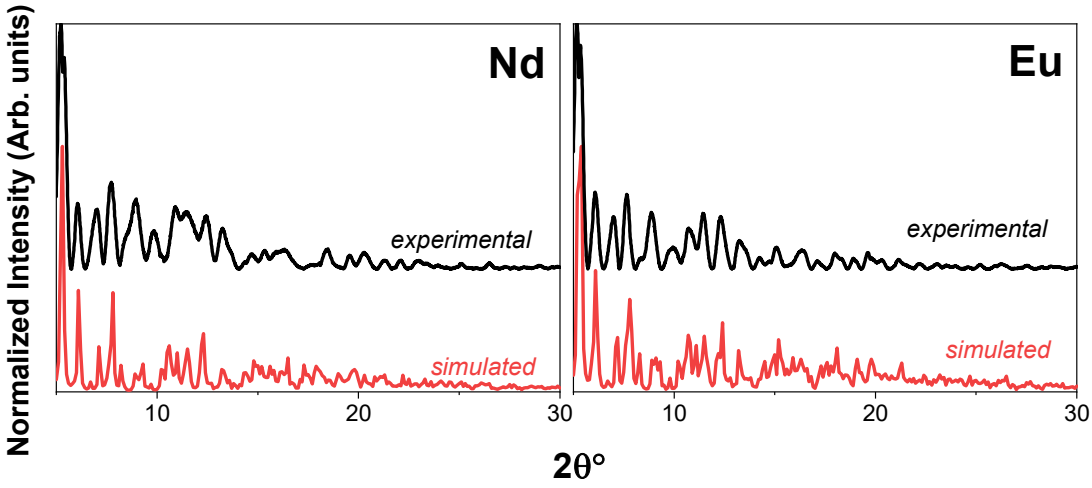

Fig. S7 Experimental PXRD pattern of **1-Nd** and **1- Eu** measured at 300 K vs simulated pattern from corresponding crystal structure measured at 100 K.

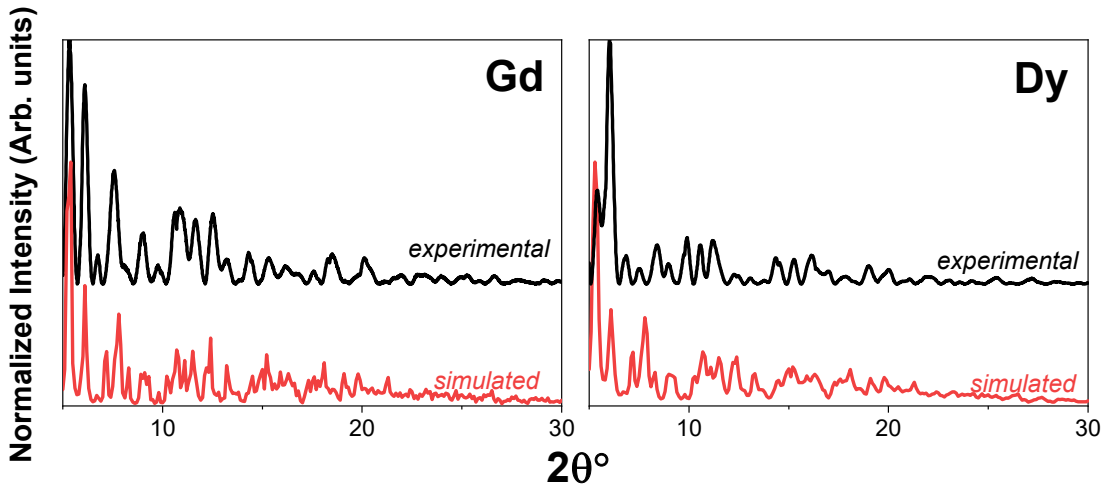

Fig. S8 Experimental PXRD pattern of **1-Gd** and **1-Dy** measured at 300 K vs simulated pattern from corresponding crystal structure measured at 100 K.

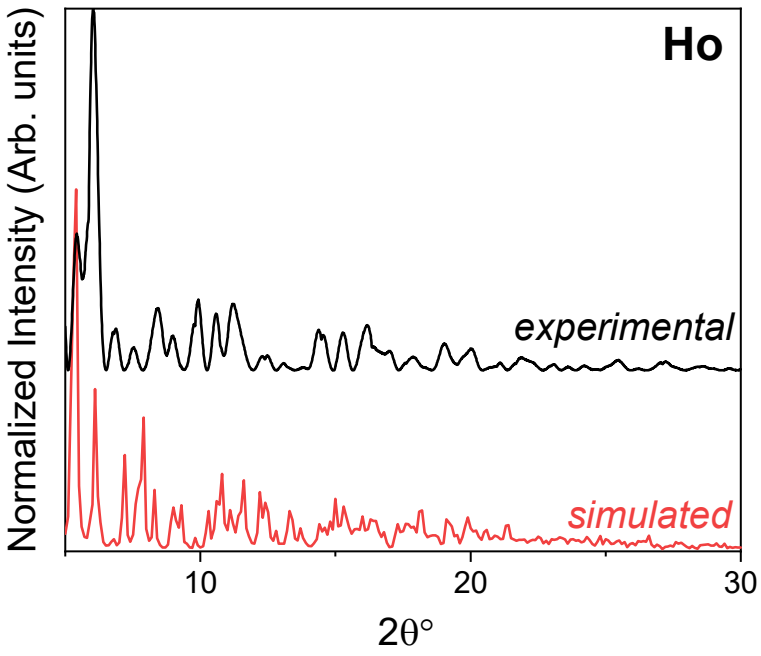

Fig. S9 Experimental PXRD pattern of **1-Ho** at 300 K vs simulated pattern from corresponding crystal structure at 100 K.

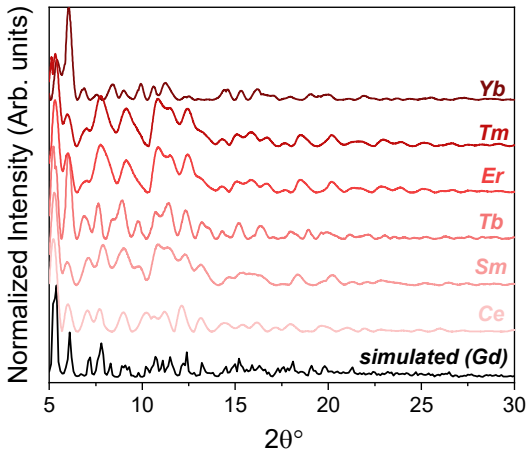

Fig. S10 1-Ln (Ln = Ce, Sm, Tb, Er, Tm, Yb) at 300 K vs simulated pattern from crystal structure of **1-Gd** measured at 100 K.

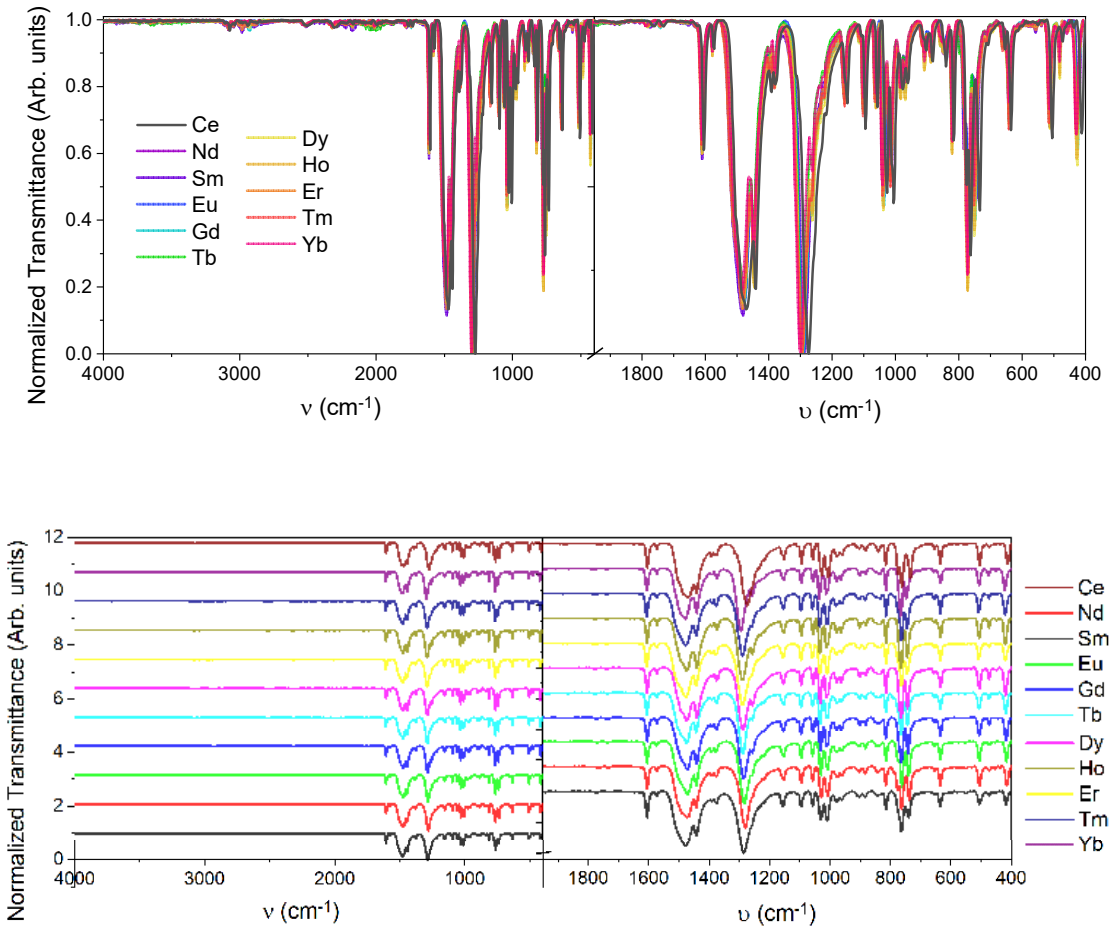

Fig. S11 Solid state infrared spectra (ATR) of compounds **1-Ln** demonstrating the similar chemical composition of the series.

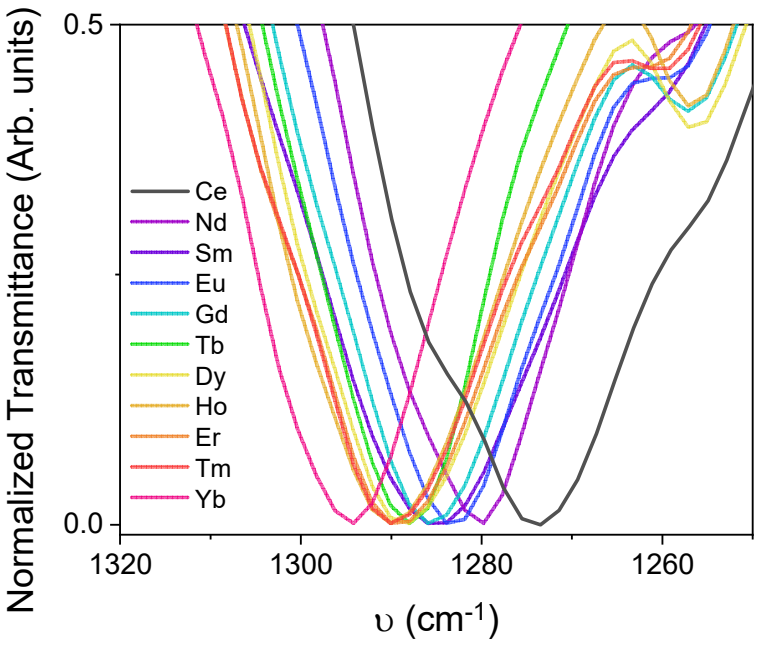

Fig. S12 Solid state infrared spectra (ATR) of compounds **1-Ln** demonstrating the demonstrating the effect of increasing mass of the lanthanide on the η^2 OONO stretch.

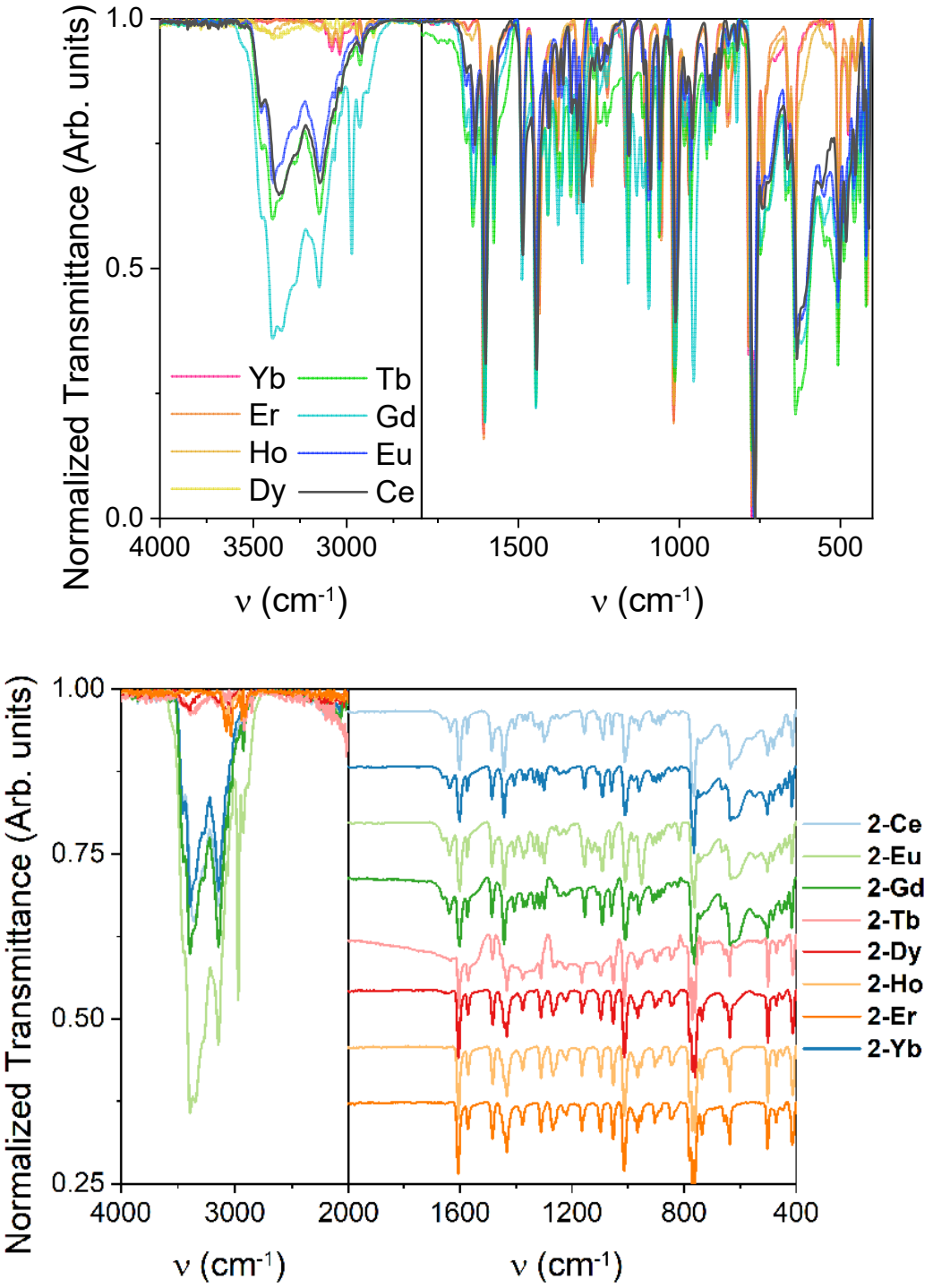

Fig. S13 Solid state infrared spectra (ATR) of compounds **2-Ln** (Ce, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb) demonstrating the similar chemical composition of the series.

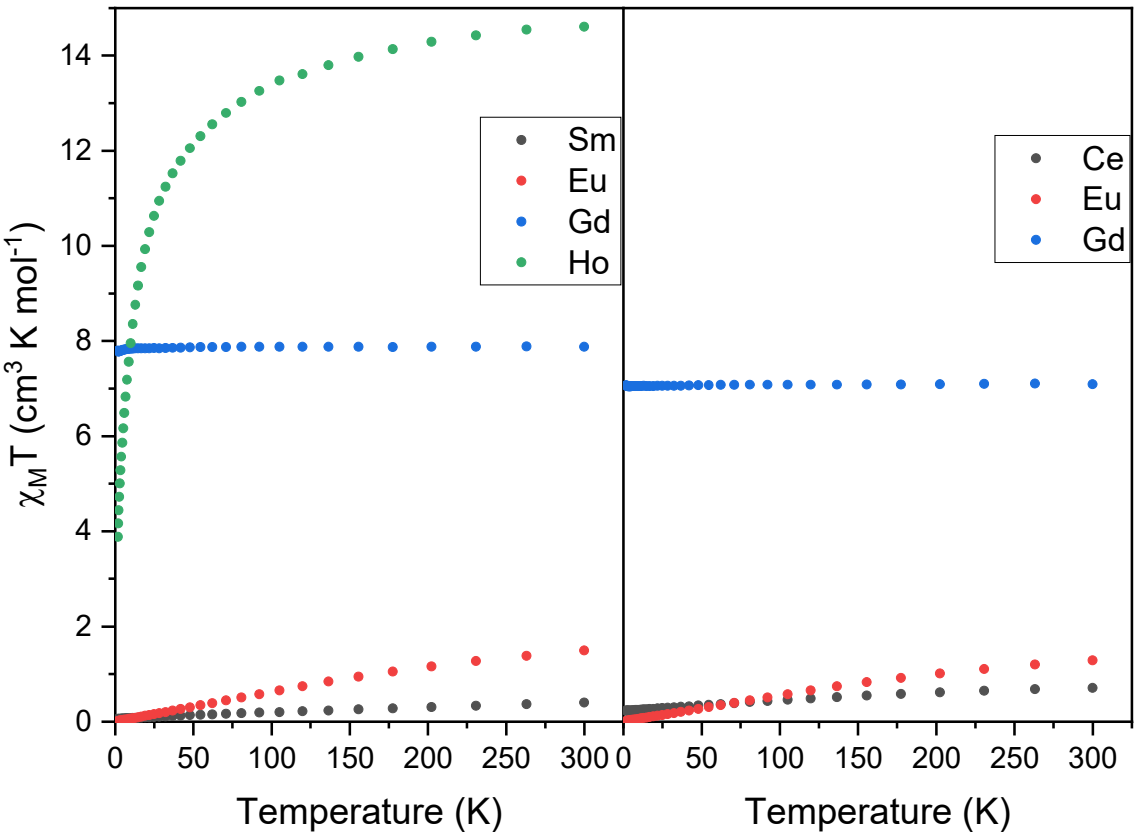

Fig. S14 Static magnetic susceptibility and temperature product in an applied field H_{dc} = 1000 Oe of complexes **1-Ln** (Ln = Sm^{3+}, Eu^{3+}, Ho^{3+}) (left), and **2-Ln** (Ln = Ce^{3+}, Eu^{3+}, Gd^{3+}) (right) in solid state.

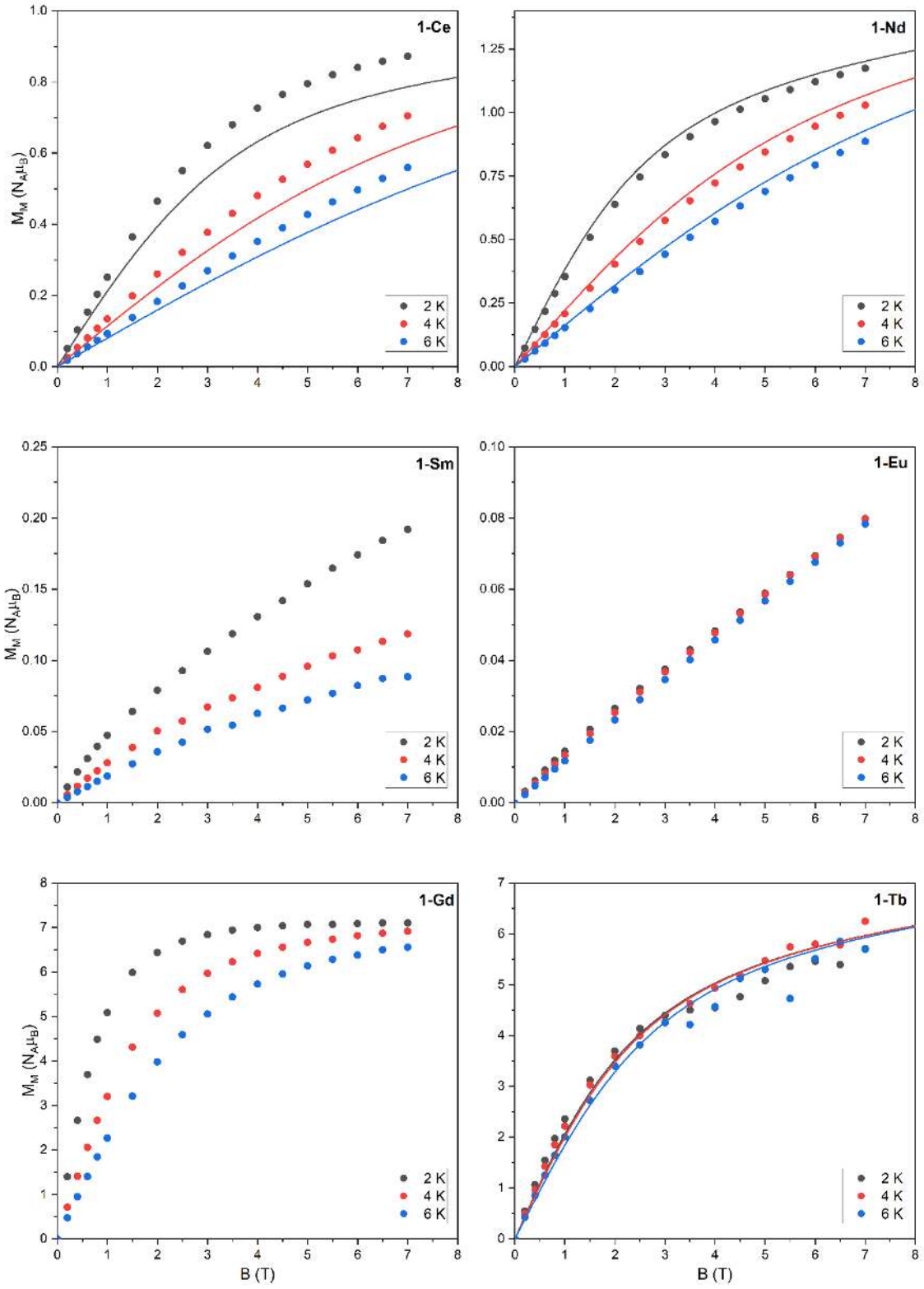

Fig. S15 Magnetisation with field for complexes **1-Ce**, **1-Nd**, **1-Sm**, **1-Eu**, **1-Gd**, and **1-Tb** in solid state. Solid lines are simulated profiles from CASSCF calculations.

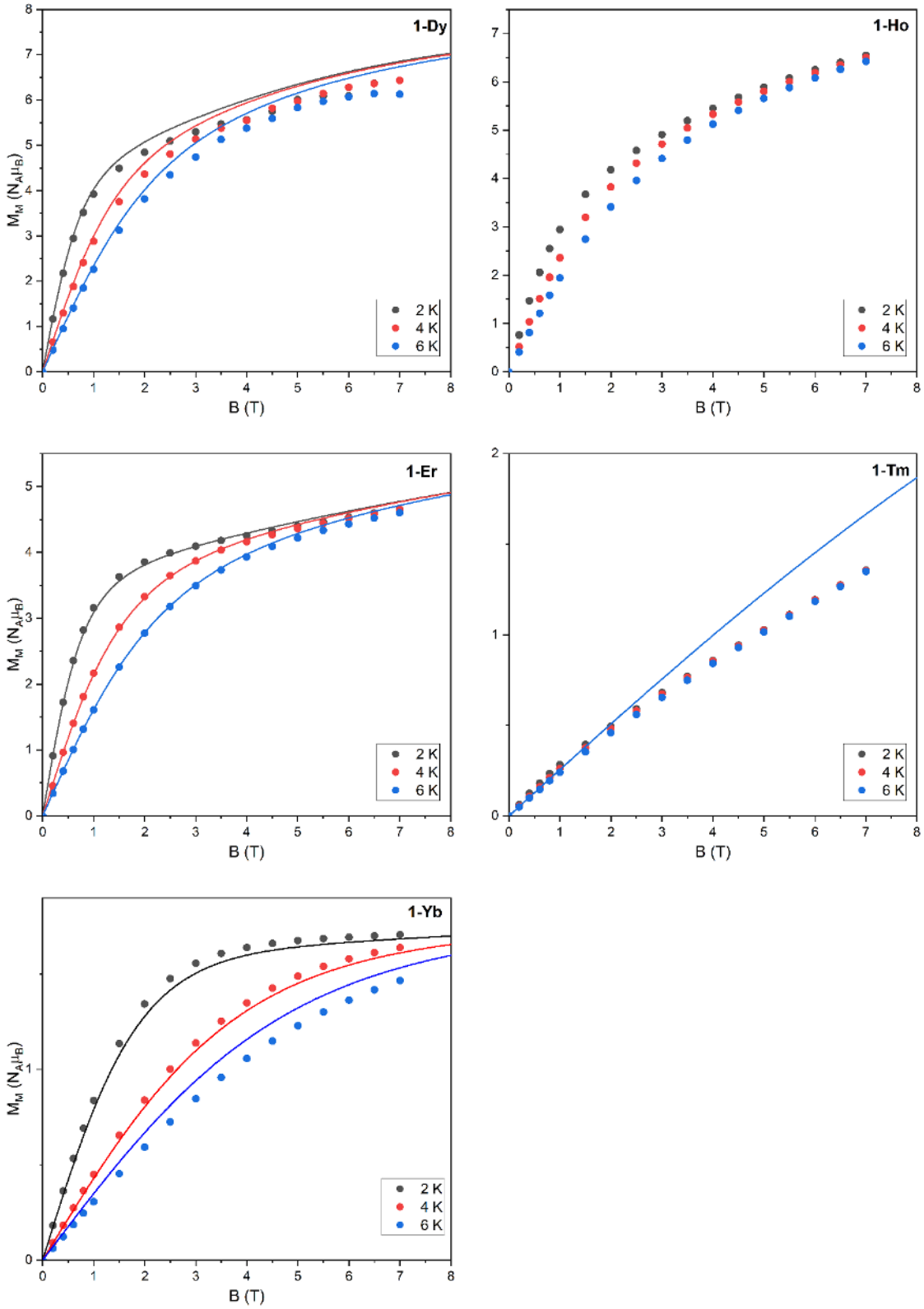

Fig. S16 Magnetisation with field for complexes **1-Dy**, **1-Ho**, **1-Er**, **1-Tm**, and **1-Yb** in solid state. Solid lines are simulated profiles from CASSCF calculations.

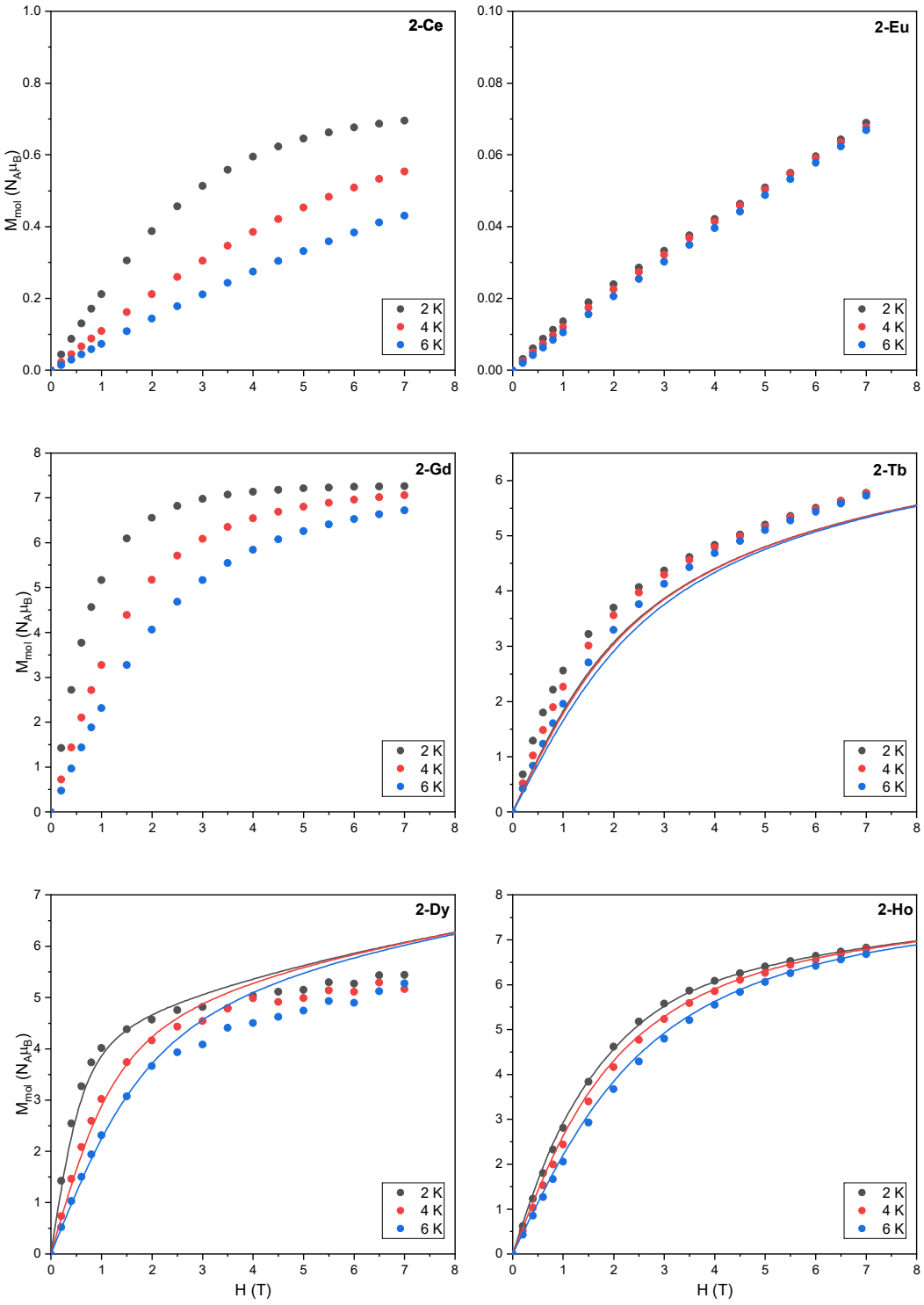

Fig. S17 Magnetisation with field for complexes **2-Ce**, **2- Eu**, **2-Gd**, **2-Tb**, **2-Dy**, and **2-Ho** in solid state. Solid lines are simulated profiles from CASSCF calculations.

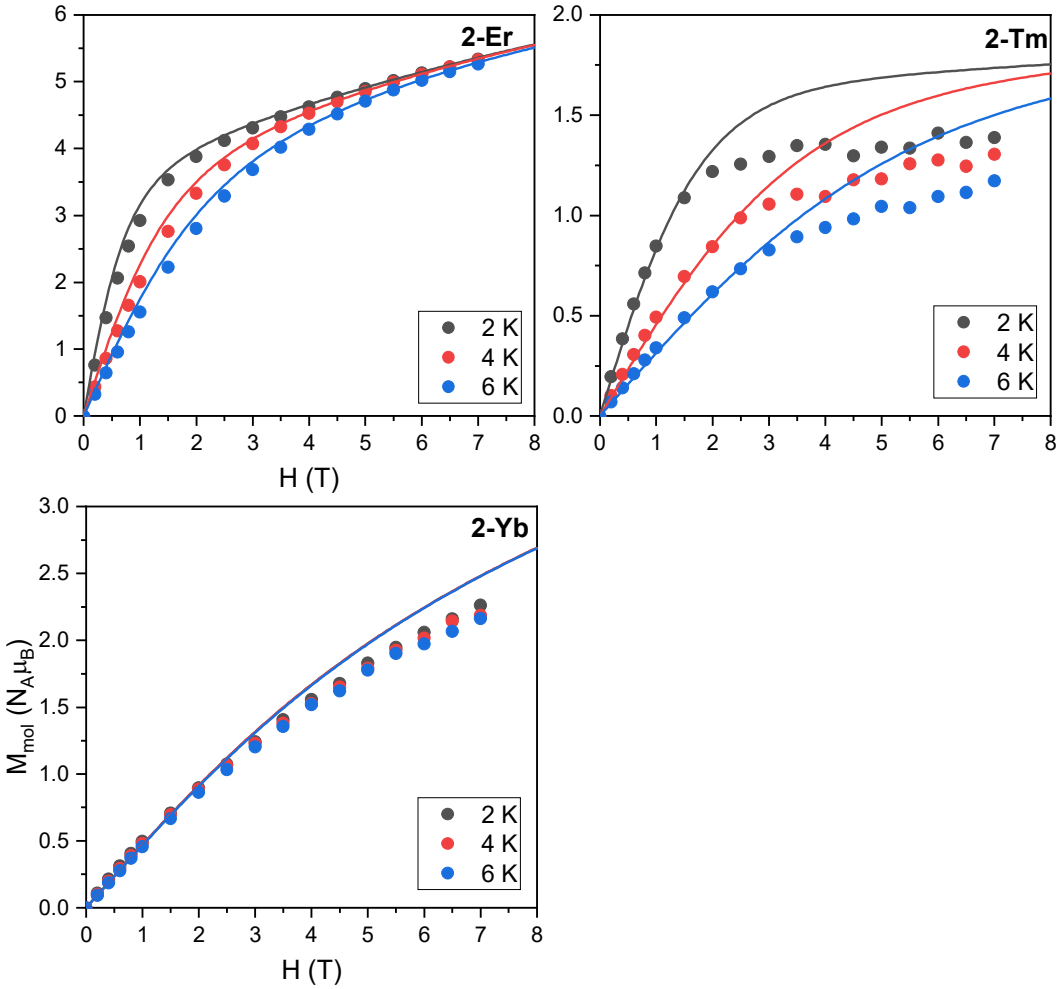

Fig. S18 Magnetisation with field for complexes **2- Er**, **2-Tm**, and **2-Yb** in solid state. Solid lines are simulated profiles from CASSCF calculations.

$$\chi' = \chi_s + (\chi_t - \chi_s)\frac{1 + (2\pi\tau\nu)^{1-\alpha}sin\left(\alpha\frac{\pi}{2}\right)}{1 + 2sin\left(\alpha\frac{\pi}{2}\right)(2\pi\tau\nu)^{1-\alpha} + (2\pi\tau\nu)^{2-2\alpha}}$$

$$\chi'' = (\chi_t - \chi_s)\frac{(2\pi\tau\nu)^{1-\alpha}cos\left(\alpha\frac{\pi}{2}\right)}{1 + 2sin\left(\alpha\frac{\pi}{2}\right)(2\pi\tau\nu)^{1-\alpha} + (2\pi\tau\nu)^{2-2\alpha}}$$

Equation S1 Double generalised Debye model for fitting of in-phase and out-of-phase magnetic susceptibility data.

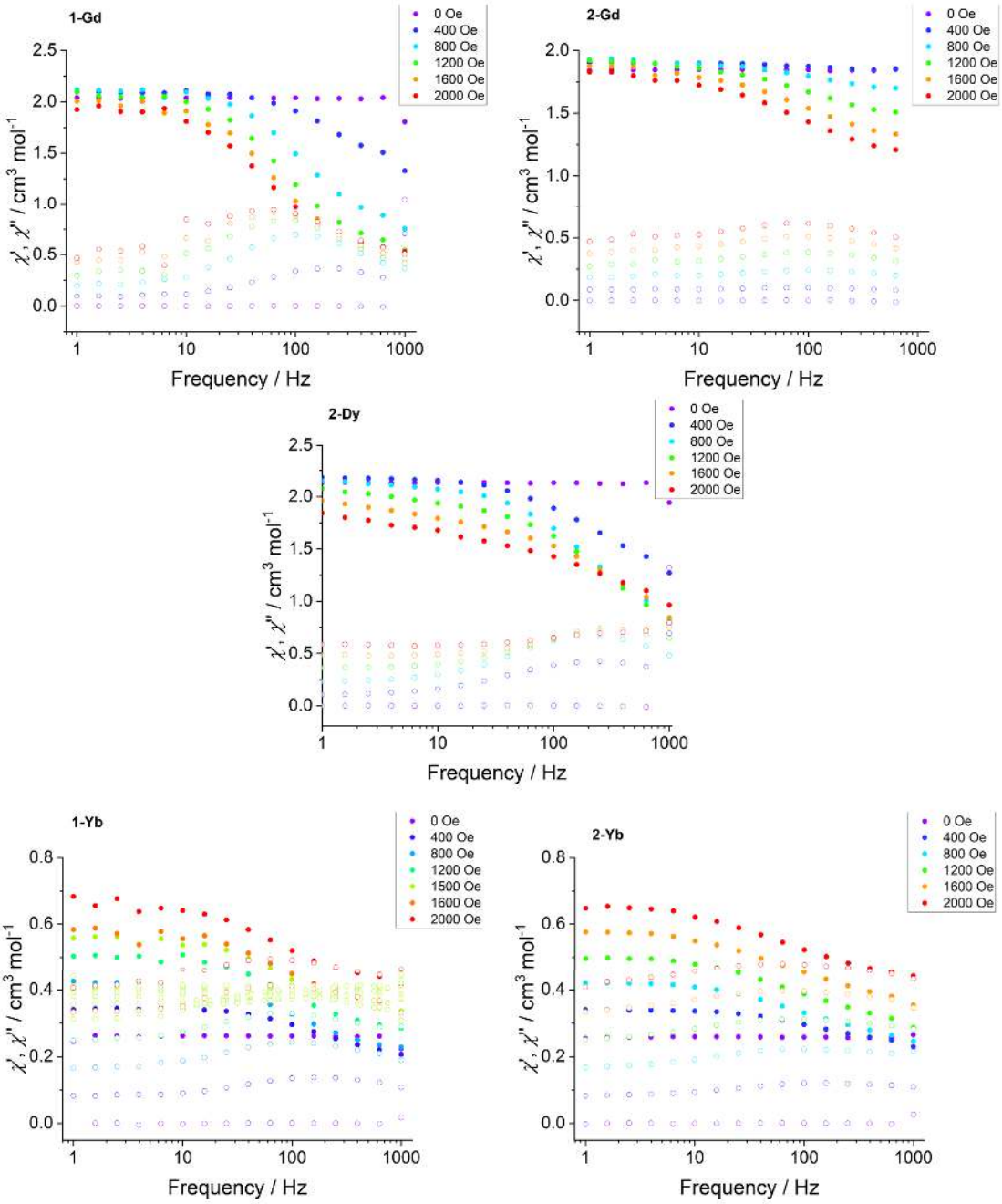

Fig. S19 In-phase and out-of-phase field dependence of the ac susceptibility of **1-Gd**, **2-Gd**, **2-Dy**, **1-Yb**, and **2-Yb** in the solid state**.**

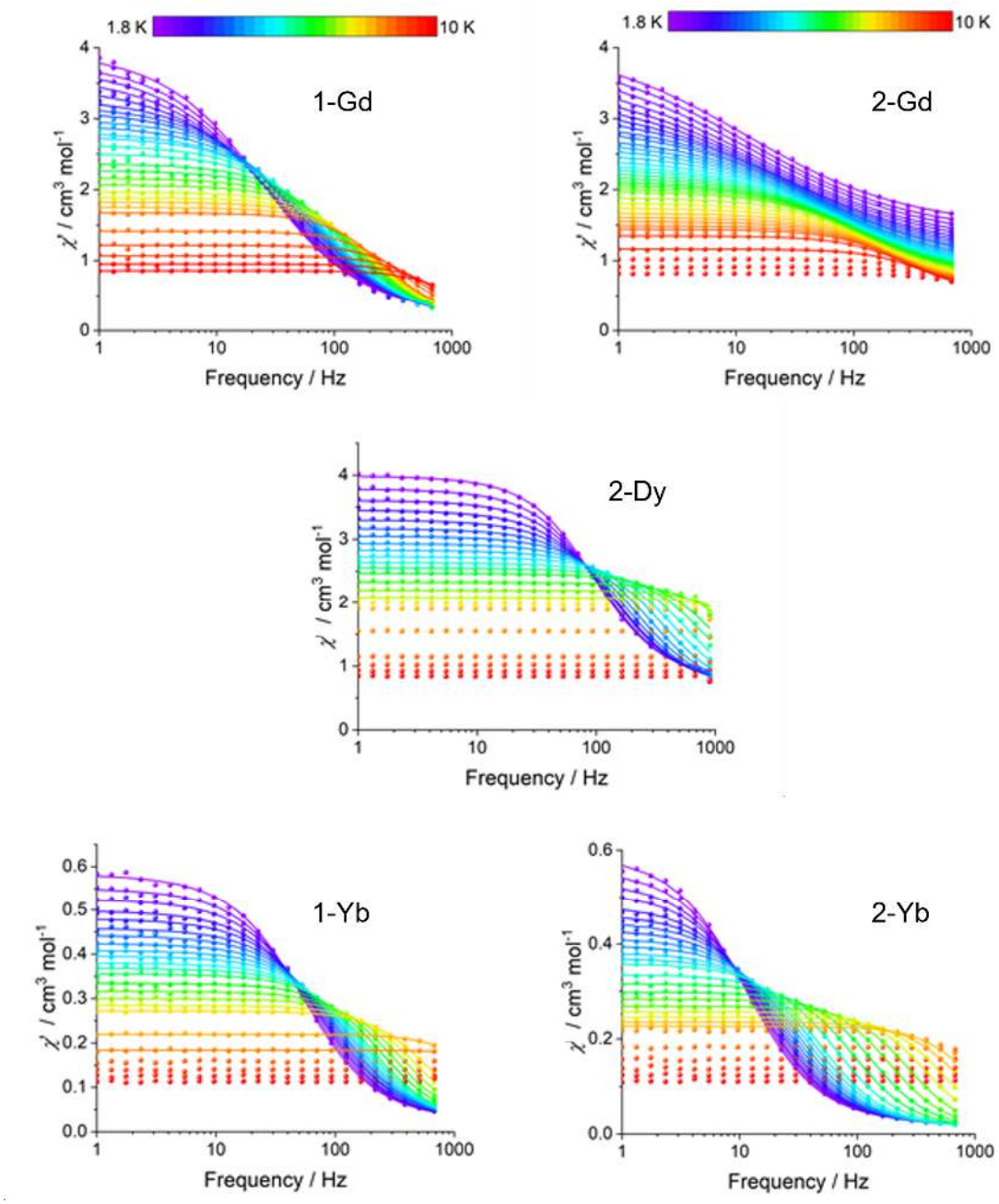

Fig. S20 In-phase component of the ac susceptibility of **1-Gd** (H_{dc} = 2000 Oe), **2-Gd** (H_{dc} = 2000 Oe), **2-Dy** (H_{dc} = 800 Oe), **1-Yb** (H_{dc} = 2000 Oe), and **2-Yb** (H_{dc} = 2000 Oe) in the solid state measured in an applied magnetic field.

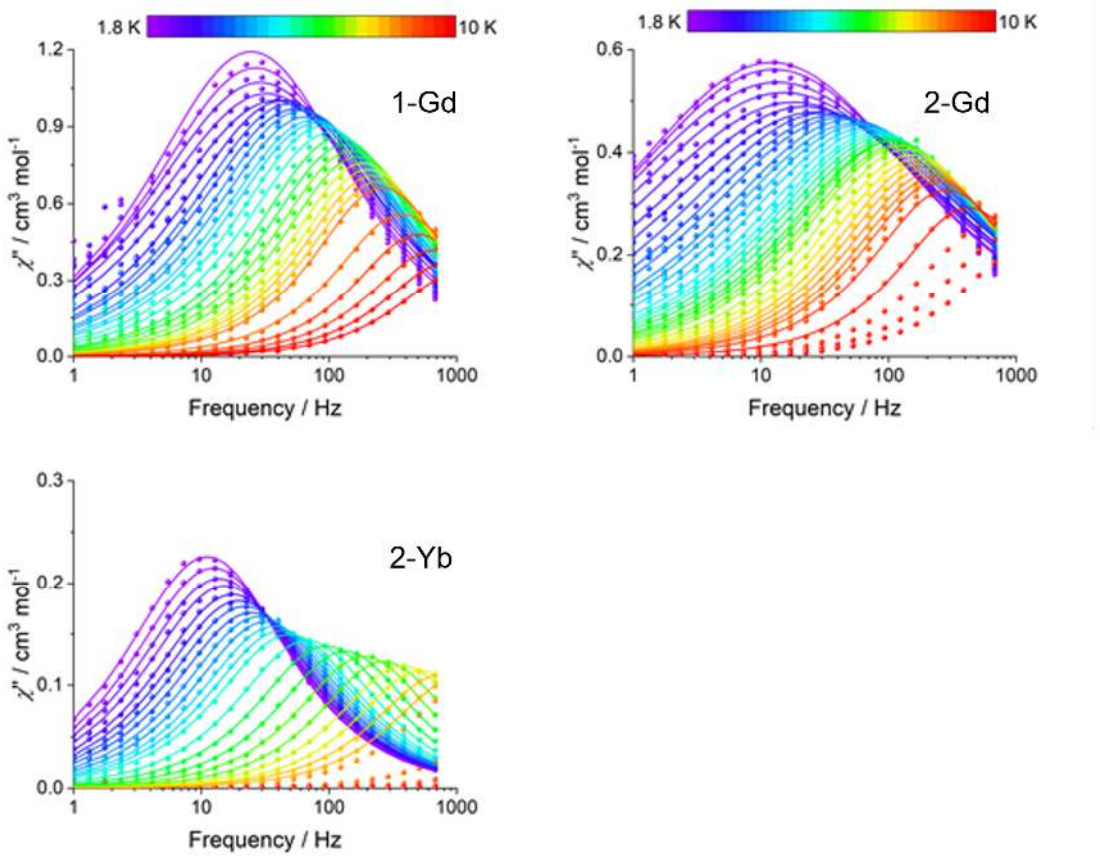

Fig. S21 Out-of-phase component of the ac susceptibility of **1-Gd** (H_{dc} = 2000 Oe), **2-Gd** (H_{dc} = 2000 Oe) and **2-Yb** (H_{dc} = 2000 Oe) in the solid state measured in an applied magnetic field.

Table S13. Fitting parameters for **2-Dy** to the generalised Debye model for temperatures where appropriate fits could be obtained.

T *(K)*	χ_s *(cm^3 mol^{-1})*	χ_t *(cm^3 mol^{-1})*	τ *(s)*	α
1.8	0.82 ± 0.01	3.989 ± 0.007	0.00167 ± 1 x 10^{-5}	0.104 ± 0.005
1.9	0.78 ± 0.01	3.785 ± 0.007	0.00152 ± 1 x 10^{-5}	0.104 ± 0.005
2	0.74 ± 0.01	3.609 ± 0.004	0.00135 ± 8 x 10^{-6}	0.109 ± 0.003
2.1	0.66 ± 0.02	3.456 ± 0.009	0.00115 ± 2 x 10^{-5}	0.126 ± 0.007
2.2	0.68 ± 0.01	3.301 ± 0.005	0.00104 ± 9 x 10^{-6}	0.110 ± 0.005
2.3	0.63 ± 0.02	3.166 ± 0.006	8.72 x 10^{-4} ± 9 x 10^{-6}	0.111 ± 0.006
2.4	0.62 ± 0.02	3.048 ± 0.005	7.29 x 10^{-4} ± 8 x 10^{-6}	0.104 ± 0.006
2.5	0.57 ± 0.02	2.933 ± 0.005	5.81 x 10^{-4} ± 8 x 10^{-6}	0.102 ± 0.007
2.6	0.54 ± 0.03	2.827 ± 0.006	4.52 x 10^{-4} ± 8 x 10^{-6}	0.098 ± 0.008
2.7	0.61 ± 0.03	2.725 ± 0.005	3.72 x 10^{-4} ± 7 x 10^{-6}	0.078 ± 0.008
2.8	0.49 ± 0.05	2.633 ± 0.006	2.67 x 10^{-4} ± 9 x 10^{-6}	0.090 ± 0.012
2.9	0.48 ± 0.05	2.549 ± 0.005	2.00 x 10^{-4} ± 8 x 10^{-6}	0.088 ± 0.012
3	0.50 ± 0.09	2.465 ± 0.007	1.55 x 10^{-4} ± 1 x 10^{-5}	0.071 ± 0.019

Table S14. Fitting parameters for **1-Gd** to the generalised Debye model for temperatures where appropriate fits could be obtained.

T *(K)*	χ_s *(cm^3 mol^{-1})*	χ_t *(cm^3 mol^{-1})*	τ *(s)*	α
1.8	0.20 ± 0.03	3.97 ± 0.03	0.0065 ± 1 x 10^{-4}	0.28 ± 0.01
1.9	0.16 ± 0.03	3.83 ± 0.03	0.0059 ± 2 x 10^{-4}	0.30 ± 0.01
2	0.12 ± 0.04	3.75 ± 0.03	0.0055 ± 2 x 10^{-4}	0.32 ± 0.01
2.1	0.09 ± 0.04	3.63 ± 0.04	0.0049 ± 2 x 10^{-4}	0.33 ± 0.02
2.2	0.08 ± 0.04	3.47 ± 0.03	0.0042 ± 1 x 10^{-4}	0.32 ± 0.01
2.3	0.12 ± 0.03	3.29 ± 0.02	0.00362 ± 9 x 10^{-5}	0.28 ± 0.01
2.4	0.11 ± 0.03	3.19 ± 0.02	0.00319 ± 9 x 10^{-5}	0.27 ± 0.01
2.5	0.09 ± 0.03	3.12 ± 0.02	0.00290 ± 9 x 10^{-5}	0.28 ± 0.01
2.6	0.11 ± 0.03	3.00 ± 0.02	0.00259 ± 7 x 10^{-5}	0.25 ± 0.01
2.7	0.10 ± 0.03	2.92 ± 0.02	0.00235 ± 6 x 10^{-5}	0.25 ± 0.01
2.8	0.11 ± 0.03	2.81 ± 0.02	0.00212 ± 5 x 10^{-5}	0.24 ± 0.01
2.9	0.11 ± 0.03	2.75 ± 0.02	0.00196 ± 5 x 10^{-5}	0.23 ± 0.01
3	0.13 ± 0.02	2.63 ± 0.01	0.00178 ± 4 x 10^{-5}	0.205 ± 0.009
3.2	0.14 ± 0.03	2.51 ± 0.01	0.00154 ± 4 x 10^{-5}	0.19 ± 0.01
3.4	0.16 ± 0.02	2.368 ± 0.008	0.00134 ± 2 x 10^{-5}	0.161 ± 0.008

3.6	0.15 ± 0.02	2.269 ± 0.009	0.00118 ± 2 x 10^{-5}	0.15 ± 0.01
3.8	0.17 ± 0.02	2.162 ± 0.008	0.00105 ± 2 x 10^{-5}	0.14 ± 0.01
4	0.18 ± 0.02	2.067 ± 0.006	0.00095 ± 2 x 10^{-5}	0.119 ± 0.008
4.2	0.19 ± 0.02	1.967 ± 0.005	0.00086 ± 1 x 10^{-5}	0.100 ± 0.008
4.4	0.20 ± 0.01	1.887 ± 0.004	0.00078 ± 1 x 10^{-5}	0.091 ± 0.007
4.6	0.20 ± 0.02	1.826 ± 0.004	0.00072 ± 1 x 10^{-5}	0.089 ± 0.008
4.8	0.20 ± 0.01	1.748 ± 0.003	0.00066 ± 8 x 10^{-6}	0.078 ± 0.006
5	0.21 ± 0.01	1.673 ± 0.003	0.00061 ± 8 x 10^{-6}	0.063 ± 0.006
6	0.21 ± 0.03	1.415 ± 0.006	0.00043 ± 2 x 10^{-5}	0.05 ± 0.02
7	0.23 ± 0.03	1.216 ± 0.003	0.00032 ± 1 x 10^{-5}	0.02 ± 0.01
8	0.21 ± 0.02	1.068 ± 0.002	0.00024 ± 9 x 10^{-6}	0.02 ± 0.01
9	0.20 ± 0.02	0.951 ± 0.001	0.00019 ± 7 x 10^{-6}	0.01 ± 0.01
10	0.19 ± 0.04	0.858 ± 0.002	0.00015 ± 1 x 10^{-5}	0.01 ± 0.02

Table S15. Fitting parameters for **2-Gd** to the generalised Debye model for temperatures where appropriate fits could be obtained.

T *(K)*	χ_s *(cm^3 mol^{-1})*	χ_t *(cm^3 mol^{-1})*	τ *(s)*	α
1.8	1.44 ± 0.01	4.088 ± 0.021	0.013 ± 3 x 10^{-4}	0.478 ± 0.007
1.9	1.35 ± 0.01	3.980 ± 0.017	0.013 ± 3 x 10^{-4}	0.486 ± 0.005
2	1.26 ± 0.02	3.873 ± 0.025	0.012 ± 4 x 10^{-4}	0.504 ± 0.008
2.1	1.19 ± 0.02	3.710 ± 0.024	0.011 ± 3 x 10^{-4}	0.505 ± 0.008
2.2	1.13 ± 0.01	3.547 ± 0.018	0.009 ± 2 x 10^{-4}	0.503 ± 0.007
2.3	1.08 ± 0.01	3.378 ± 0.013	0.007 ± 1 x 10^{-4}	0.487 ± 0.005
2.4	1.03 ± 0.01	3.253 ± 0.015	0.006 ± 1 x 10^{-4}	0.483 ± 0.007
2.5	1.00 ± 0.01	3.117 ± 0.011	0.005 ± 1 x 10^{-4}	0.461 ± 0.006
2.6	0.96 ± 0.02	2.995 ± 0.014	0.004 ± 1 x 10^{-4}	0.446 ± 0.008
2.7	0.94 ± 0.02	2.908 ± 0.012	0.004 ± 1 x 10^{-4}	0.437 ± 0.008
2.8	0.91 ± 0.01	2.779 ± 0.010	0.0033 ± 8 x 10^{-5}	0.415 ± 0.007
2.9	0.87 ± 0.01	2.705 ± 0.009	0.0029 ± 7 x 10^{-5}	0.412 ± 0.007
3	0.85 ± 0.01	2.617 ± 0.007	0.0027 ± 5 x 10^{-5}	0.396 ± 0.007
3.1	0.83 ± 0.01	2.532 ± 0.008	0.0024 ± 6 x 10^{-5}	0.384 ± 0.007
3.2	0.82 ± 0.02	2.436 ± 0.009	0.0022 ± 6 x 10^{-5}	0.360 ± 0.009
3.3	0.80 ± 0.01	2.366 ± 0.006	0.0020 ± 4 x 10^{-5}	0.351 ± 0.007
3.4	0.78 ± 0.01	2.312 ± 0.004	0.0019 ± 3 x 10^{-5}	0.344 ± 0.005
3.5	0.76 ± 0.01	2.250 ± 0.004	0.0017 ± 3 x 10^{-5}	0.334 ± 0.006

3.6	0.75 ± 0.02	2.190 ± 0.007	0.0016 ± 4 x 10^{-5}	0.321 ± 0.009
3.7	0.75 ± 0.01	2.130 ± 0.004	0.0015 ± 2 x 10^{-5}	0.304 ± 0.006
3.8	0.74 ± 0.01	2.081 ± 0.005	0.0015 ± 3 x 10^{-5}	0.287 ± 0.008
3.9	0.73 ± 0.01	2.030 ± 0.004	0.0014 ± 2 x 10^{-5}	0.279 ± 0.006
4	0.72 ± 0.01	1.981 ± 0.004	0.0013 ± 2 x 10^{-5}	0.263 ± 0.006
4.2	0.71 ± 0.01	1.892 ± 0.003	0.0012 ± 2 x 10^{-5}	0.237 ± 0.006
4.4	0.68 ± 0.01	1.813 ± 0.003	0.0011 ± 2 x 10^{-5}	0.223 ± 0.006
4.6	0.67 ± 0.01	1.741 ± 0.003	0.0010 ± 2 x 10^{-5}	0.207 ± 0.007
4.8	0.66 ± 0.01	1.670 ± 0.003	0.0009 ± 2 x 10^{-5}	0.184 ± 0.008
5	0.64 ± 0.01	1.609 ± 0.003	0.0008 ± 1 x 10^{-5}	0.175 ± 0.007
5.2	0.62 ± 0.01	1.552 ± 0.002	0.0008 ± 1 x 10^{-5}	0.163 ± 0.008
5.4	0.61 ± 0.01	1.493 ± 0.002	0.0007 ± 1 x 10^{-5}	0.142 ± 0.008
5.6	0.60 ± 0.01	1.444 ± 0.002	0.0007 ± 1 x 10^{-5}	0.129 ± 0.008
6	0.57 ± 0.01	1.350 ± 0.002	0.0006 ± 1 x 10^{-5}	0.108 ± 0.008
7	0.51 ± 0.01	1.162 ± 0.001	0.00038 ± 7 x 10^{-6}	0.066 ± 0.008

Table S16. Fitting parameters for **1-Yb** to the generalised Debye model for temperatures where appropriate fits could be obtained.

T *(K)*	χ_s *(cm^3 mol^{-1})*	χ_t *(cm^3 mol^{-1})*	τ *(s)*	α
1.8	0.024 ± 0.004	0.5830 ± 0.0026	0.00300 ± 5 x 10^{-5}	0.147 ± 0.009
1.9	0.024 ± 0.001	0.5512 ± 0.0009	0.00265 ± 2 x 10^{-5}	0.144 ± 0.003
2	0.022 ± 0.002	0.5259 ± 0.0011	0.00235 ± 2 x 10^{-5}	0.143 ± 0.004
2.1	0.021 ± 0.002	0.4993 ± 0.0009	0.00209 ± 2 x 10^{-5}	0.141 ± 0.004
2.2	0.023 ± 0.003	0.4807 ± 0.0015	0.00190 ± 2 x 10^{-5}	0.138 ± 0.007
2.3	0.021 ± 0.002	0.4589 ± 0.0009	0.00168 ± 1 x 10^{-5}	0.130 ± 0.004
2.4	0.021 ± 0.002	0.4426 ± 0.0009	0.00152 ± 1 x 10^{-5}	0.127 ± 0.005
2.5	0.022 ± 0.001	0.4242 ± 0.0005	0.00136 ± 8 x 10^{-6}	0.116 ± 0.003
2.6	0.022 ± 0.002	0.4085 ± 0.0009	0.00122 ± 1 x 10^{-5}	0.109 ± 0.006
2.7	0.021 ± 0.003	0.3956 ± 0.0011	0.00110 ± 2 x 10^{-5}	0.108 ± 0.007
2.8	0.021 ± 0.002	0.3800 ± 0.0006	0.00099 ± 8 x 10^{-6}	0.099 ± 0.004
2.9	0.022 ± 0.002	0.3680 ± 0.0005	0.00090 ± 7 x 10^{-6}	0.091 ± 0.004
3	0.022 ± 0.002	0.3555 ± 0.0005	0.00080 ± 7 x 10^{-6}	0.082 ± 0.004
3.2	0.024 ± 0.002	0.3343 ± 0.0004	0.00066 ± 5 x 10^{-6}	0.068 ± 0.004
3.4	0.022 ± 0.002	0.3161 ± 0.0005	0.00053 ± 6 x 10^{-6}	0.061 ± 0.005
3.6	0.025 ± 0.002	0.2996 ± 0.0004	0.00043 ± 5 x 10^{-6}	0.046 ± 0.006

3.8	0.022 ± 0.004	0.2842 ± 0.0006	0.00034 ± 7 x 10^{-6}	0.042 ± 0.009
4	0.018 ± 0.005	0.2719 ± 0.0005	0.00026 ± 7 x 10^{-6}	0.048 ± 0.010

Table S17. Fitting parameters for **2-Yb** to the generalised Debye model for temperatures where appropriate fits could be obtained.

T *(K)*	χ_s *(cm^3 mol^{-1})*	χ_t *(cm^3 mol^{-1})*	τ *(s)*	α
1.8	0.019 ± 0.001	0.594 ± 0.002	0.01432 ± 1 x 10^{-4}	0.151 ± 0.004
1.9	0.019 ± 0.001	0.563 ± 0.002	0.01278 ± 8 x 10^{-5}	0.148 ± 0.004
2	0.0169 ± 8 x 10^{-4}	0.536 ± 0.001	0.11490 ± 6 x 10^{-5}	0.151 ± 0.003
2.1	0.0167 ± 7 x 10^{-4}	0.5126 ± 9 x 10^{-4}	0.01037 ± 5 x 10^{-5}	0.144 ± 0.002
2.2	0.0161 ± 8 x 10^{-4}	0.484 ± 0.001	0.00904 ± 5 x 10^{-5}	0.137 ± 0.003
2.3	0.016 ± 0.001	0.465 ± 0.001	0.00811 ± 6 x 10^{-5}	0.131 ± 0.004
2.4	0.015 ± 0.001	0.448 ± 0.001	0.00727 ± 5 x 10^{-5}	0.128 ± 0.004
2.5	0.0143 ± 7 x 10^{-4}	0.4311 ± 7 x 10^{-4}	0.00644 ± 3 x 10^{-5}	0.126 ± 0.002
2.6	0.015 ± 0.001	0.413 ± 0.001	0.00565 ± 4 x 10^{-5}	0.113 ± 0.004
2.7	0.0142 ± 5 x 10^{-4}	0.3977 ± 5 x 10^{-4}	0.00497 ± 2 x 10^{-5}	0.107 ± 0.002
2.8	0.014 ± 0.001	0.3814 ± 5 x 10^{-4}	0.00431 ± 3 x 10^{-5}	0.099 ± 0.004
2.9	0.0129 ± 8 x 10^{-4}	0.3701 ± 6 x 10^{-4}	0.00376 ± 2 x 10^{-5}	0.098 ± 0.003
3	0.0123 ± 8 x 10^{-4}	0.3593 ± 6 x 10^{-4}	0.00325 ± 2 x 10^{-5}	0.094 ± 0.003
3.2	0.0127 ± 9 x 10^{-4}	0.3352 ± 5 x 10^{-4}	0.00236 ± 1 x 10^{-5}	0.074 ± 0.003
3.4	0.012 ± 0.001	0.3162 ± 6 x 10^{-4}	0.00169 ± 1 x 10^{-5}	0.060 ± 0.004
3.6	0.0121 ± 8 x 10^{-4}	0.2984 ± 4 x 10^{-4}	0.00119 ± 6 x 10^{-6}	0.046 ± 0.003
3.8	0.0114 ± 9 x 10^{-4}	0.2839 ± 3 x 10^{-4}	8.41 x 10^{-4} ± 4 x 10^{-6}	0.039 ± 0.003
4	0.011 ± 0.001	0.2689 ± 4 x 10^{-4}	5.91 x 10^{-4} ± 5 x 10^{-6}	0.030 ± 0.004
4.2	0.010 ± 0.002	0.2572 ± 4 x 10^{-4}	4.21 x 10^{-4} ± 5 x 10^{-6}	0.027 ± 0.005
4.4	0.011 ± 0.003	0.2458 ± 4 x 10^{-4}	3.07 x 10^{-4} ± 6 x 10^{-6}	0.019 ± 0.008
4.6	0.000 ± 0.006	0.2360 ± 4 x 10^{-4}	2.11 x 10^{-4} ± 7 x 10^{-6}	0.041 ± 0.010
4.8	0.004 ± 0.006	0.2265 ± 3 x 10^{-4}	1.59 x 10^{-4} ± 6 x 10^{-6}	0.028 ± 0.009

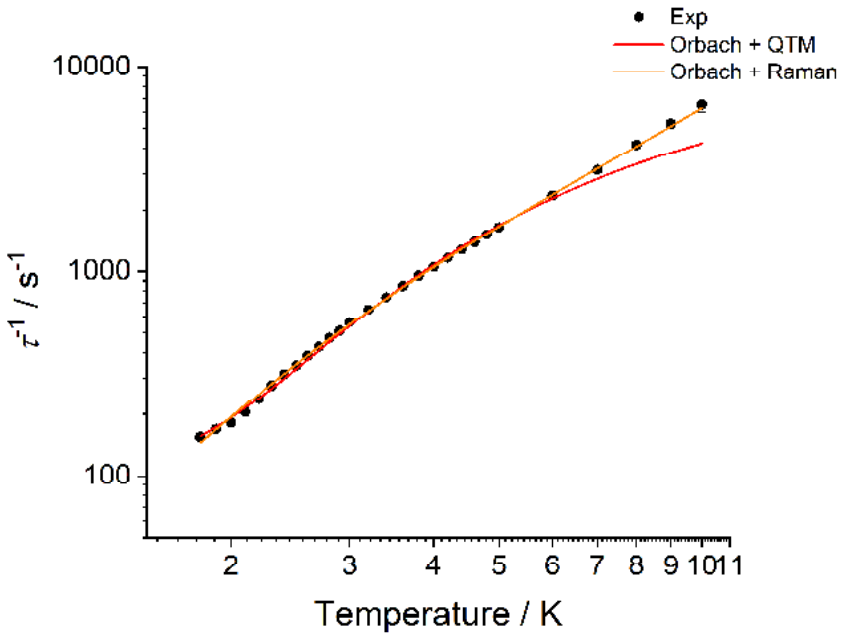

Fig. S22 Fitting of the in-field AC relaxation rates of **1-Gd** with QTM and Raman terms.

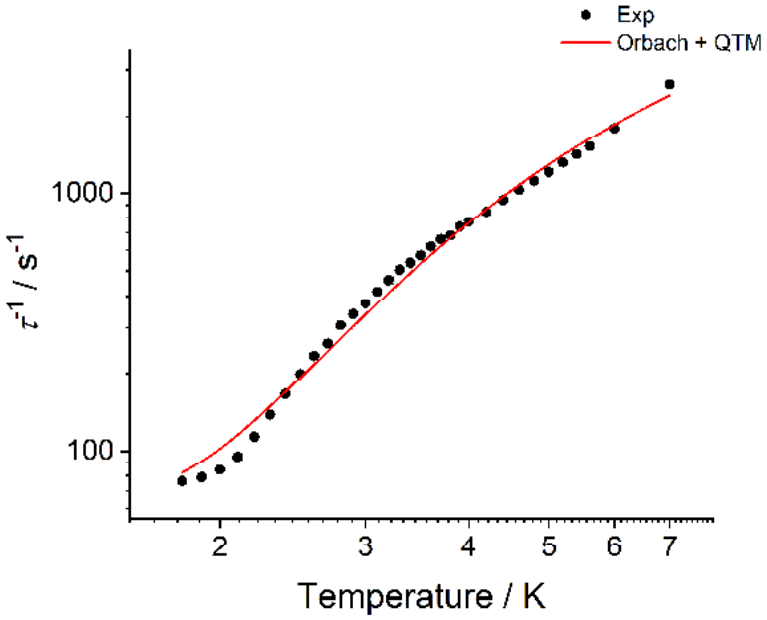

Fig. S23 Fitting of the in-field AC relaxation rates of **2-Gd** with QTM and Raman terms.

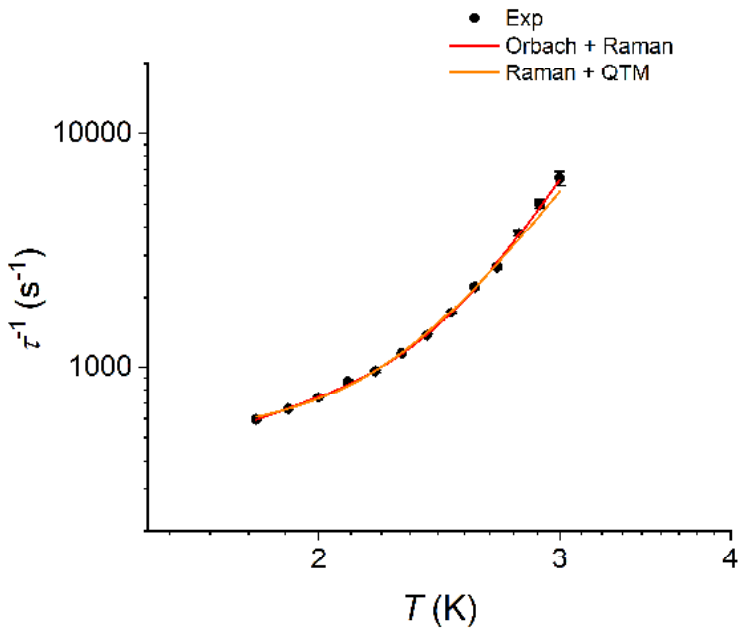

Fig. S24 Fitting of the in-field AC relaxation rates of **2-Dy** with QTM and Raman terms.

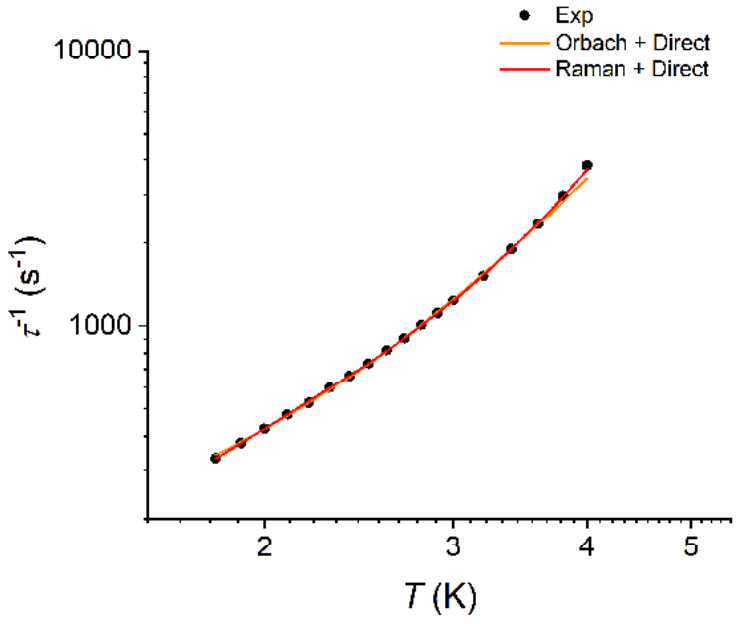

Fig. S25 Fitting of the in-field AC relaxation rates of **1-Yb** with QTM and Raman terms.

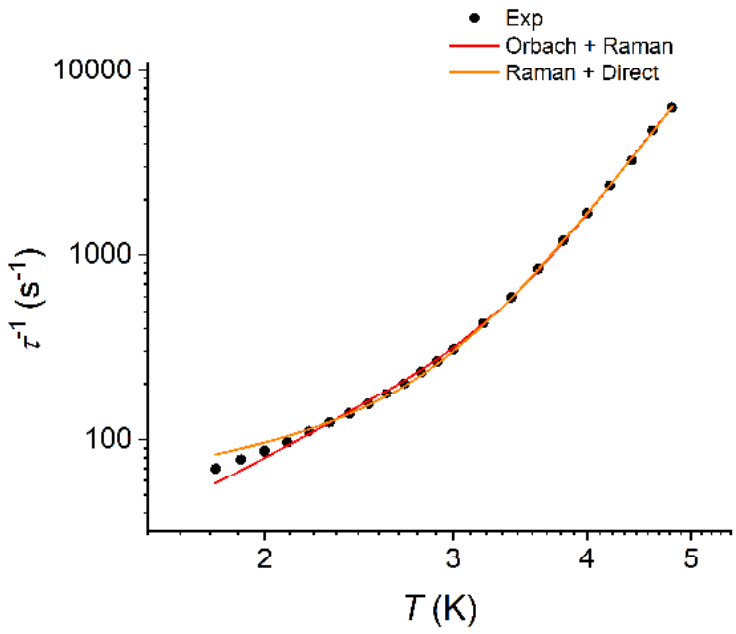

Fig. S26 Fitting of the in-field AC relaxation rates of **2-Yb** with QTM and Raman terms.

Table S18 CASSCF energies, g-tensor values, g_z angle and wavefunctions for the lowest-lying states of **1-Ce.**

Energy (cm^{-1})	g_x	g_y	g_z	Angle	Wavefunction
0.0	0.7	1.3	2.3	--	54%|∓5/2⟩, 40%|±1/2⟩
69.4	0.5	0.9	2.5	41	93%|±3/2⟩
223.6	1.2	1.5	1.6	61	43%|±5/2⟩, 57%|∓1/2⟩

Table S19 CASSCF energies, g-tensor values, g_z angle and wavefunctions for the lowest-lying states of **1-Nd**.

Energy (cm^{-1})	g_x	g_y	g_z	Angle	Wavefunction
0.0	0.6	1.3	3.3	--	40%|∓9/2⟩, 49%|∓3/2⟩
23.0	0.5	1.2	2.7	78	13%|±7/2⟩, 61%|∓5/2⟩, 15%|±1/2⟩
178	1.9	2.6	2.7	52	38%|±7/2⟩, 53%|±1/2⟩
282	0.2	0.5	3.9	26	52%|∓9/2⟩, 44%|±3/2⟩
436	1.6	2.8	3.2	89	46%|±7/2⟩, 24%|±5/2⟩, 29%|∓1/2⟩

Table S20 CASSCF energies, g-tensor values, g_z angle and wavefunctions for the lowest-lying states of **1-Dy**.

Energy (cm^{-1})	g_x	g_y	g_z	Angle	Wavefunction
0.0	3.6	7.7	10.6	--	33%|±13/2⟩, 20%|±7/2⟩, 21%|±1/2⟩
10.7	1.0	2.7	13.7	69	45%|∓15/2⟩, 28%|∓9/2⟩, 12%|∓3/2⟩
60.0	0.2	2.4	8.1	70	23%|±13/2⟩, 23%|∓11/2⟩, 13%|∓5/2⟩, 27%|∓1/2⟩
91.0	2.1	3.4	8.4	51	19%|∓15/2⟩, 59%|∓3/2⟩
115	0.3	1.0	12.4	49	13%|±13/2⟩, 10%|∓7/2⟩, 32%|±5/2⟩, 27%|±1/2⟩
158	5.0	5.5	8.0	39	10%|±13/2⟩, 55%|∓11/2⟩, 16%|±5/2⟩, 11%|±1/2⟩
223	4.4	7.2	9.6	18	15%|±15/2⟩, 45%|±9/2⟩, 16%|∓7/2⟩, 10%|∓3/2⟩
241	0.2	1.2	15.2	53	19%|∓9/2⟩, 38%|±7/2⟩, 20%|∓5/2⟩

Table S21 CASSCF energies, g-tensor values, g_z angle and wavefunctions for the lowest-lying states of **1- Er**.

Energy (cm^{-1})	g_x	g_y	g_z	Angle	Wavefunction
0.0	3.4	4.7	11.2	--	15%|∓13/2⟩, 26%|∓7/2⟩, 13%|±5/2⟩, 35%|∓1/2⟩
44.3	2.7	3.7	7.8	88	15%|±15/2⟩, 44%|±9/2⟩, 30%|±3/2⟩
132	3.9	4.7	6.3	71	16%|∓13/2⟩, 58%|±11/2⟩
164	2.7	5.8	7.6	89	60%|∓13/2⟩, 13%|∓11/2⟩, 20%|±5/2⟩
244	0.8	1.5	9.4	88	47%|∓15/2⟩, 40%|±3/2⟩
282	2.2	5.7	8.1	47	17%|∓11/2⟩, 18%|±7/2⟩, 27%|∓5/2⟩, 30%|±1/2⟩,
362	2.2	4.8	9.8	35	42%|±7/2⟩, 22%|∓5/2⟩, 19%|±1/2⟩
385	2.5	3.7	10.2	84	33%|∓15/2⟩, 41%|∓9/2⟩, 18%|±3/2⟩

Table S22 CASSCF energies and wavefunctions for the lowest-lying states of **1-Tb**.

Energy (cm^{-1})	-6	-5	-4	-3	-2	-1	0	1	2	3	4	5	6
0	13	3	1	29	2	0	5	0	2	29	1	3	13
9.82	0	13	20	1	12	5	0	5	12	1	20	13	0
12.6	12	11	5	12	9	1	1	1	9	12	5	11	12
22.1	1	17	20	1	4	6	0	6	4	1	20	17	1
39.6	13	3	15	10	8	1	1	1	8	10	15	3	13
44.2	1	17	16	6	3	7	0	7	3	6	16	17	1
83.3	25	2	0	4	5	0	26	0	5	4	0	2	25
100	1	5	9	0	15	20	0	20	15	0	9	5	1
101	8	9	3	9	9	13	0	13	9	9	3	9	8
112	8	7	5	5	9	16	1	16	9	5	5	7	8
116	11	10	2	10	14	1	4	1	14	10	2	10	11
125	6	0	3	5	4	10	44	10	4	5	3	0	6
125	2	5	1	8	5	20	18	20	5	8	1	5	2

Table S23 CASSCF energies, g-tensor values, g_z angle and wavefunctions for the lowest-lying states of **1-Yb**.

ENERGY (CM^{-1})	G_X	G_Y	G_Z	**ANGLE**	**WAVEFUNCTION**
0.0	2.5	3.2	3.8	--	41%|±7/2⟩, 17%|∓5/2⟩, 18%|±3/2⟩, 23%|∓1/2⟩
1**04**	0.2	0.4	5.2	11	23%|∓7/2⟩, 46%|∓5/2⟩, 13%|∓3/2⟩, 18%|∓1/2⟩
146	0.4	2.8	3.8	59	23%|∓7/2⟩, 28%|∓5/2⟩, 33%|±3/2⟩, 15%|∓1/2⟩,
215	0.8	1.3	3.2	76	12%|∓7/2⟩, 10%|±5/2⟩, 35%|±3/2⟩, 44%|±1/2⟩

Table S24 CASSCF energies and wavefunctions for the lowest-lying states of **1-Tm**.

Energy (cm^{-1})	-6	-5	-4	-3	-2	-1	0	1	2	3	4	5	6
0	8	0	0	37	1	0	5	0	1	37	0	0	8
48.9	0	2	21	2	22	3	0	3	22	2	21	2	0
86.1	0	6	18	0	20	6	0	6	20	0	18	6	0
103	21	0	1	27	1	0	1	0	1	27	1	0	21
145	0	18	7	0	3	21	2	21	3	0	7	18	0
148	2	16	10	1	12	10	0	10	12	1	10	16	2
187	3	9	13	2	4	5	28	5	4	2	13	9	3
194	8	2	5	1	3	15	36	15	3	1	5	2	8
211	0	3	18	0	10	17	3	17	10	0	18	3	0
266	34	1	1	10	0	3	2	3	0	10	1	1	34

270	1	20	5	0	15	8	2	8	15	0	5	20	1
282	3	21	2	1	8	12	6	12	8	1	2	21	3
332	21	1	1	19	1	1	15	1	1	19	1	1	21

Table S25 CASSCF energies, g-tensor values, g_z angle and wavefunctions for the lowest-lying states of **2-Dy**.

Energy (cm^{-1})	g_x	g_y	g_z	Angle	Wavefunction
0.0	2.4	5.3	13.5	--	56%|∓13/2⟩, 23%|∓7/2⟩
34.1	1.6	3.5	11.4	23	41%|∓11/2⟩, 10%|∓9/2⟩, 24%|∓5/2⟩, 10%|∓3/2⟩
53.6	0.2	2.6	14.4	89	10%|±11/2⟩, 23%|∓9/2⟩, 27%|∓3/2⟩, 21%|∓1/2⟩
86.9	3.0	6.8	11.4	84	12%|±13/2⟩, 10%|∓11/2⟩, 15%|±3/2⟩, 52%|±1/2⟩
178	0.2	1.4	13.5	31	55%|∓15/2⟩, 26%|∓3/2⟩
271	1.6	4.0	13.0	70	24%|∓11/2⟩, 16%|±7/2⟩, 37%|∓5/2⟩
296	2.2	3.4	12.7	23	13%|±9/2⟩, 41%|±7/2⟩, 14%|∓5/2⟩,
322	0.4	1.6	15.7	56	23%|∓15/2⟩, 34%|∓9/2⟩, 13%|∓7/2⟩, 13%|∓3/2⟩

Table S26 CASSCF energies, g-tensor values, g_z angle and wavefunctions for the lowest-lying states of **2-Er**.

Energy (cm^{-1})	g_x	g_y	g_z	Angle	Wavefunction
0.0	3.1	4.0	11.4	--	71%|∓13/2⟩, 23%|∓1/2⟩
19.4	4.9	5.1	7.2	77	20%|∓13/2⟩, 12%|∓7/2⟩, 53%|∓1/2⟩
56	0.4	3.3	8.1	49	18%|∓11/2⟩, 22%|∓9/2⟩, 54%|∓3/2⟩
71	1.6	2.2	12.5	47	50%|∓11/2⟩, 22%|±5/2⟩, 14%|±3/2⟩
222	0.1	0.1	17.1	6	92%|±15/2⟩
256	3.8	5.9	7.7	74	19%|±11/2⟩, 12%|±7/2⟩, 42%|∓5/2⟩, 15%|±1/2⟩
290	0.2	3.7	8.3	11	63%|±9/2⟩, 24%|±3/2⟩
311	3.7	5.3	9.3	85	67%|∓7/2⟩, 22%|±5/2⟩

Table S27 CASSCF energies, g-tensor values, g_z angle and wavefunctions for the lowest-lying states of **2-Yb**.

Energy (cm^{-1})	g_x	g_y	g_z	Angle	Wavefunction
0.0	2.1	3.3	4.1	--	33%|∓7/2⟩, 19%|±5/2⟩, 47%|∓1/2⟩
179	0.3	2.8	3.6	27	19%|∓7/2⟩, 69%|±5/2⟩
248	0.8	1.4	3.2	83	82%|±3/2⟩
313	2.0	3.8	4.2	43	41%|±7/2⟩, 10%|±3/2⟩, 44%|±1/2⟩

Table S28 CASSCF energies and wavefunctions for the lowest-lying states of **2-Tb**

Energy (cm^{-1})	-6	-5	-4	-3	-2	-1	0	1	2	3	4	5	6
0	10	2	2	19	3	3	20	3	3	19	2	2	10
10.4	1	7	8	2	15	15	2	15	15	2	8	7	1
22.4	14	5	3	18	7	3	1	3	7	18	3	5	14
38.3	2	12	11	1	19	5	1	5	19	1	11	12	2
57.0	11	3	8	7	4	17	2	17	4	7	8	3	11
67.5	2	9	12	2	11	13	4	13	11	2	12	9	2
113	25	1	0	0	0	2	42	2	0	0	0	1	25
181	21	1	8	17	2	2	0	2	2	17	8	1	21
208	3	17	9	7	5	7	1	7	5	7	9	17	3
213	4	2	23	3	1	16	0	16	1	3	23	2	4
234	4	16	2	12	8	2	10	2	8	12	2	16	4
257	2	13	4	8	12	5	11	5	12	8	4	13	2
261	0	13	9	3	12	9	6	9	12	3	9	13	0

Table S29 CASSCF energies and wavefunctions for the lowest-lying states of **2-Ho**

Energy (cm^{-1})	g_z	-8	-7	-6	-5	-4	-3	-2	-1	0	1	2	3	4	5	6	7	8
0	-	0	1	0	1	36	2	6	4	0	4	6	2	36	1	0	1	0
5.21	11.3	0	0	2	3	16	21	7	0	0	0	7	21	16	3	2	0	0
6.54		0	0	1	3	19	22	3	1	0	1	3	22	19	3	1	0	0
29.9	7.5	2	0	0	25	8	0	6	9	0	9	6	0	8	25	0	0	2
35.1		2	0	1	29	1	3	10	3	2	3	10	3	1	29	1	0	2
58.3	-	2	0	9	1	1	26	2	1	15	1	2	26	1	1	9	0	2
80.0	19	47	0	0	2	0	0	0	0	0	0	0	0	0	2	0	0	47
81.3		46	0	0	1	1	1	0	0	1	0	0	1	1	1	0	0	46
201	9.3	0	2	3	14	1	5	25	1	0	1	25	5	1	14	3	2	0
208		0	0	6	13	1	2	25	2	0	2	25	2	1	13	6	0	0
219	12.3	0	3	34	4	1	2	1	4	0	4	1	2	1	4	34	3	0
236		0	2	28	2	2	8	6	2	1	2	6	8	2	2	28	2	0
252	4.5	0	2	6	0	6	2	7	27	1	27	7	2	6	0	6	2	0
257		0	2	1	1	7	1	1	36	3	36	1	1	7	1	1	2	0
284	-	0	8	6	0	0	3	0	1	63	1	0	3	0	0	6	8	0
287	14	0	43	1	0	0	1	0	3	2	3	0	1	0	0	1	43	0
289		0	35	1	0	0	2	0	6	12	6	0	2	0	0	1	35	0

Table S30 CASSCF energies and wavefunctions for the lowest-lying states of **2-Tm**

Energy (cm^{-1})	g_z	-6	-5	-4	-3	-2	-1	0	1	2	3	4	5	6
0	9.7	17	0	0	31	1	1	0	1	1	31	0	0	17
21.0		21	1	1	20	3	1	6	1	3	20	1	1	21
63.1	5.7	2	7	7	3	30	1	0	1	30	3	7	7	2
84.3		0	11	4	1	32	1	0	1	32	1	4	11	0
100	7.5	1	0	30	1	1	16	1	16	1	1	30	0	1
118		0	5	22	2	5	15	2	15	5	2	22	5	0
194	9.6	30	1	2	15	1	1	1	1	1	15	2	1	30
207		23	1	0	12	1	4	18	4	1	12	0	1	23
264	-	1	0	18	0	4	23	8	23	4	0	18	0	1
276	-	3	2	1	13	1	2	57	2	1	13	1	2	3
280	-	0	0	15	0	2	32	2	32	2	0	15	0	0
315	9.7	1	35	0	1	9	1	3	1	9	1	0	35	1
323		1	35	0	1	12	1	1	1	12	1	0	35	1

Table S31 Fitting parameters for the 77 K emission data of **1-Yb** using Voigt profiles with shared Gaussian and Lorentzian widths.

	Peak 1	Peak 2	Peak 3	Peak 4
y_0	0.0138 ± 8.78 x 10^{-4}	0.0138 ± 8.78 x 10^{-4}	0.0138 ± 8.78 x 10^{-4}	0.013 ± 8.78 x 10^{-4}
x_c	974.679 ± 0.051	990.23 ± 0.32	1000.87 ± 0.085	1017.15 ± 0.086
A	9.00 ± 0.16	2.18 ± 0.37	15.40 ± 0.52	10.69 ± 0.27
w_G	6.48 ± 0.36			
w_L	5.47 ± 0.35			
Reduced χ^2	3.89 x 10^{-4}			
R^2 (COD)	0.98731			
Adj. R^2	0.98706			

Table S32 Fitting parameters for the 77 K emission data of **2-Yb** using Voigt profiles with shared Gaussian and Lorentzian widths.

	Peak 1	Peak 2	Peak 3	Peak 4
y_0	0.030 ± 0.0010	0.0303 ± 0.001	0.0303 ± 0.0010	0.0304 ± 0.0010
x_c	973.39 ± 0.11	1005.58 ± 0.15	1016.49 ± 0.073	1033.35 ± 0.06
A	6.50 ± 0.13	5.71 ± 0.13	11.70 ± 0.16	10.85 ± 0.14
w_G	6.48218 ± 0.35788			
w_L	5.47572 ± 0.35141			
Reduced χ^2	4.97783 x 10^{-4}			
R^2 (COD)	0.98271			
Adj. R^2	0.98242			

$$I = A_0 e^{-\frac{t}{\tau}} + I_0$$

Equation S2 Monoexponential model used for the fitting of the decay data for **1-Ln** and **2-Ln.**

Table S33 Monoexponential fitting parameters for the decay data for **1-Ln** and **2-Ln** (Ln = Eu, Tb, Dy, Yb) at room temperature.

	τ (ns)	A_0 (Arb. Units)	I_0 (Arb. Units)
1- Eu	1261(6)·10^3	0.895(2)	23(8) ·10^{-4}
2- Eu	337.0(7)·10^3	0.935(1)	10(3)·10^{-4}
1-Tb	1653(10)·10^3	0.906(3)	0.003(1)
2-Tb	2157(17)·10^3	0.797(3)	0.005(1)
1-Dy	29.681(55) ·10^3	1.030(1)	-8(2)·10^{-4}
2-Dy	9.521(20) ·10^3	1.055(2)	-3(1)·10^{-4}
1-Yb	11.031(17) ·10^3	1.047(1)	9(1)·10^{-4}
2-Yb	8.161(11) ·10^3	1.059(1)	9.1(8)·10^{-4}

Table S34 Monoexponential fitting parameters for the decay data for **1-Ln** and **2-Ln** (Ln = Eu, Tb, Dy, Yb) at 77 K.

	τ (ns)	**A_0 (Arb. Units)**	**I_o (Arb. Units)**
1- Eu	$1310(7)\cdot10^3$	0.895(2)	$23(8)\cdot10^{-4}$
2- Eu	$339.2(1)\cdot10^3$	0.935(1)	$10(3)\cdot10^{-4}$
1-Tb	$1487(11)\cdot10^3$	0.906(3)	0.003(1)
2-Tb	$2378(23)\cdot10^3$	0.797(3)	0.005(1)
1-Dy	$27.25(6)\cdot10^3$	1.030(1)	$-8(2)\cdot10^{-4}$
2-Dy	$8.58(1)\cdot10^3$	1.055(2)	$-3(1)\cdot10^{-4}$
1-Yb	$11.03(2)\cdot10^3$	1.047(1)	$9(1)\cdot10^{-4}$
2-Yb	$7.086(8)\cdot10^3$	1.059(1)	$9.1(8)\cdot10^{-4}$

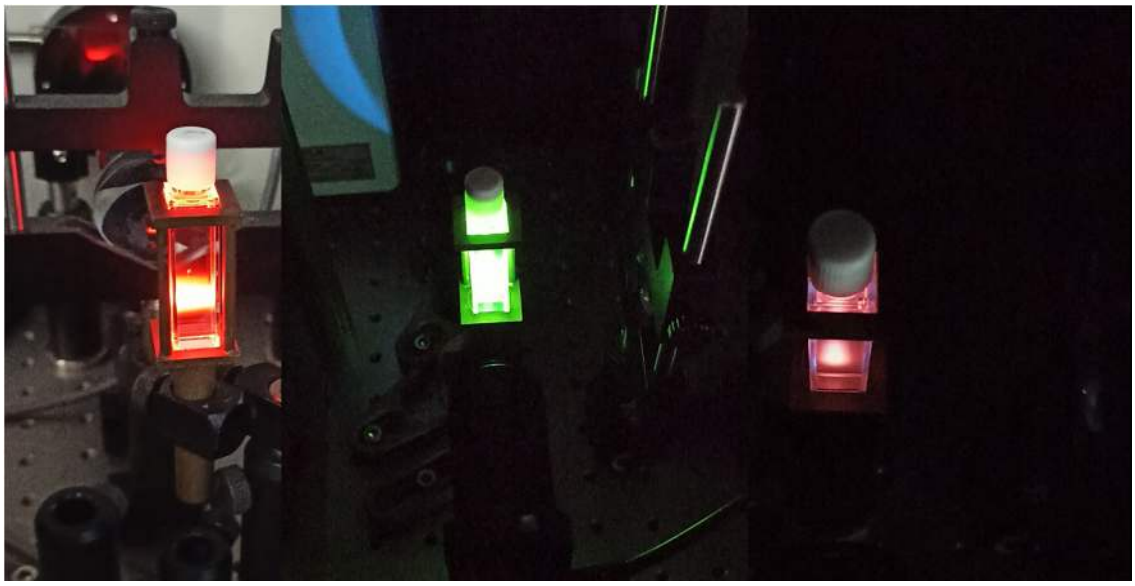

Fig. S27 Emission in the visible region by compounds **1-Eu** (left), **1-Tb** (centre), and **1-Dy** (right).

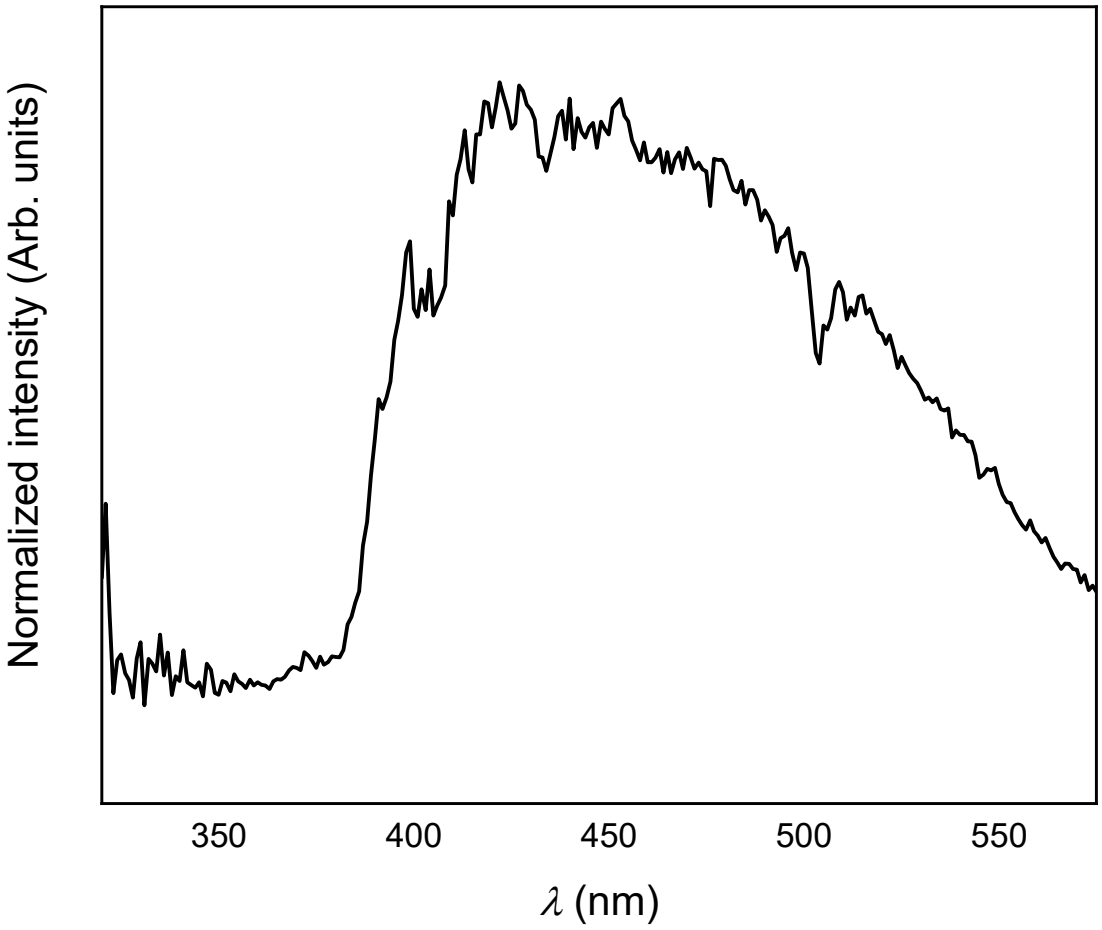

Figure S28 Emission spectrum of polycrystalline powder samples of **1-Gd** when excited at 280 nm.

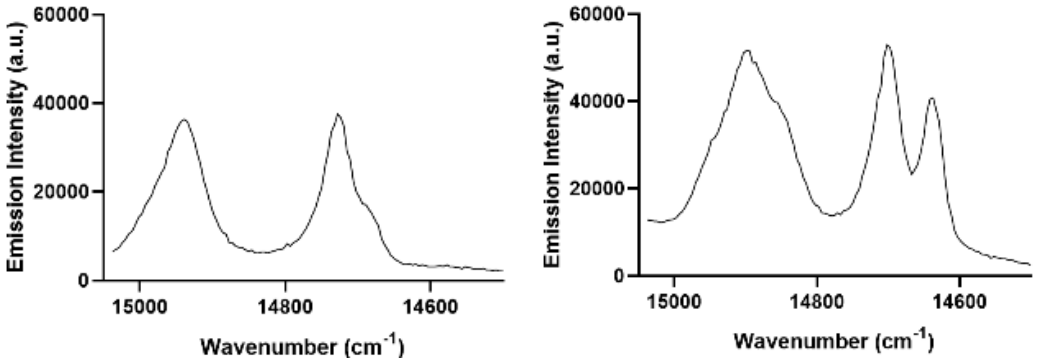

Figure S29 $^5D_4 \rightarrow {}^7F_0$ transition of **1-Tb** (left) and **2-Tb** (right).

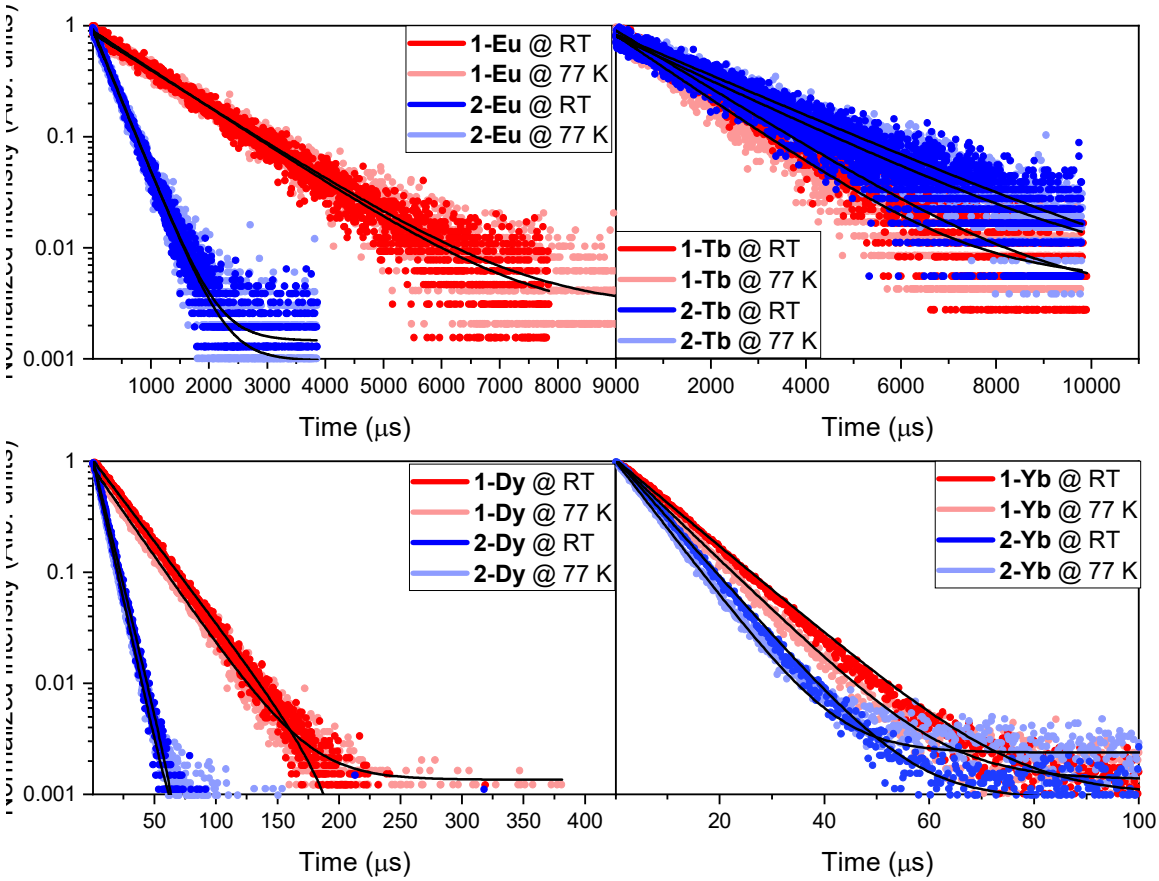

Fig. S30 Normalised time-resolved photoluminescence data for compounds **1-Ln**, and **2-Ln** (Ln = Eu, Tb, Dy, and Yb) monitored at the maximum emission wavelength($^5D_0 \rightarrow {}^7F_2$ for **1-Eu** and **2-Eu** (615 nm), $^5D_4 \rightarrow {}^7F_5$ for **1-Tb** and **2-Tb** (545 nm), $^4F_{9/2} \rightarrow {}^6H_{13/2}$ for **1-Dy** and **2-Dy** (575 nm), and $^2F_{5/2} \rightarrow {}^2F_{7/2}$ for **1-Yb** and **2-Yb** (980 nm). Solid lines indicate the single exponential fitting of the decay curves.